

A Study on Optogalvanic-like Effect in Discharge and Vector Representation of Transition-Probability

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(Received September 21, 2004)

Abstract: We report the observation of a sudden but transient dip in discharge current in presence of a strong laser radiation. This behaviour is being explained in the light of Bloch¹ equations for explaining matter-radiation interaction. The solution of Bloch equations by Rabi's technique² under the consideration of various broadening factors in the medium indicates to the nutation of the Bloch vectors at a frequency

$$\Omega(\Delta) \cong \sqrt{\Delta^2 + (K\epsilon_0)^2} .$$
 As it can be seen from the calculations of Rabi that the

nutation frequency Ω is a function of the detuning Δ . Taking into account of this factor we have been able to explain the observed current dip in terms of dephasing of different groups of molecules. We categorise the whole medium into different groups of molecules according to their frequency difference with the applied laser field (detuning Δ). As it is evident from the Rabi's solutions that these groups undergo nutation at their own frequencies under the influence of the laser field thereby leading to complete dephasing among the groups as far as their transition probability is concerned. This dephasing is the key factor behind the current dip in the discharge tube. The variation in transition probability is being explained using a unique way in which the peaks of each transition probability curve are joined to form vectors whose orientations with respect to a particular direction indicate the state of the molecules of the medium as far as their transition to an excited state is concerned.

1. Introduction

Laser induced changes in discharge current has already been reported in the form of Optogalvanic effect³. This work is slightly different from the above-mentioned phenomenon as here the laser is being applied only at the positive column of the discharge whereas in the Optogalvanic effect the cathode of the tube is exposed to laser. Also in this work the decrease in discharge current is being explained in terms of optical nutation^{4,5} of the molecular dipoles.

It is always convenient to study the response of a molecular medium to an external optical field by considering it as a two-level molecular system. This assumption is an essential precondition for our study on resonance and near resonance phenomena arising

out of interaction of laser with matter. Our system consists of CO molecules present near the positive column in a discharge operating near threshold and the intense electric field is provided by the 100 mw Ar⁺ laser.

Extension of Bloch's work to optical region leads us to a set of equations⁵ showing the behavior of the Bloch vectors and hence the population density in presence of the laser field. These set of equations, when solved according to Rabi's² and Torrey's⁶ prescriptions lead to the fact that the dipole vectors undergo nutation about the field vector and thereby induces periodic variation in the population density in the excited state. This in turn takes the on-resonance molecules to off-resonance state periodically and this is the key to periodic variation of rate of ionisation of the molecules. However this is implied only to those molecules, which are at near resonance with the applied field. It is also observed that the quick loss in periodicity of nutation can be explained in terms of various decay times associated with the dipoles.

2. Nutation of the dipole vectors

The dynamic response of a two level system to the applied field is governed by the Liouville equation for density matrix ρ^* i.e.

$$(1) \quad i\hbar \frac{d\rho^*}{dt} = [H_0 + H_{int}, \rho^*] + i\hbar \left(\frac{d\rho^*}{dt} \right)_{damping}$$

Also in terms of density matrix, the expected values of the electric dipole vector's components are given by

$$(2a) \quad \langle \rho_x \rangle = \frac{\gamma(\rho_{-+}^* + \rho_{+-}^*)}{\sqrt{2}},$$

$$(2b) \quad \langle \rho_y \rangle = \frac{\gamma(\rho_{-+}^* - \rho_{+-}^*)}{i\sqrt{2}},$$

and

$$(2c) \quad \langle \rho_z \rangle = \gamma(\rho_{++}^* - \rho_{--}^*),$$

Now in presence of an electric field the equation of motion of the dipole vectors become

$$(3) \quad \frac{d}{dt} \langle \vec{\rho} \rangle = -\frac{\gamma}{\hbar} \vec{E}_{eff} \times \langle \vec{\rho} \rangle - \frac{1}{T_2} (\hat{x}\rho_x + \hat{y}\rho_y) - \frac{1}{T_1} \hat{z}(\rho_z - \rho_z^0).$$

In the above equation the 2nd and 3rd terms of RHS are relaxation terms representing various decaying factors present in the medium. These terms do not play any significant role in controlling the response of the molecules to the applied field. So if we neglect the relaxation terms appearing in equation (3), it becomes analogous to the Bloch equation¹

$$(4) \quad \frac{d\vec{M}}{dt} = \gamma [\vec{M} \times \vec{H}]$$

representing the variation of nuclear magnetic polarisation vector $\vec{M}$ with time in presence of an external field vector $\vec{H}$. Where γ is the gyromagnetic ratio. Similarly equation (3) gives the variation of the molecules' dipole moment vector in presence of an optical field vector $\vec{F}$ as

$$(5) \quad \frac{\partial \vec{\rho}}{\partial t} = \vec{F} \times \vec{\rho}$$

Here $\vec{\rho}$ has three components $\bar{\rho}_1, \bar{\rho}_2$ and $\bar{\rho}_3$. The third component gives inversion, i.e. the population density in the excited level. Equation (5) shows the analogy between Bloch's work and the density matrix formalism.

Solution of Bloch's equations taking account of various decay times associated with the dipoles in the medium lead us to a new set of equations:

$$(6a) \quad \frac{\partial \bar{\rho}_1}{\partial t} = -\Delta \rho_2 - \frac{\rho_1}{T_2^{\downarrow}}$$

$$(6b) \quad \frac{\partial \bar{\rho}_2}{\partial t} = \Delta \rho_1 - \frac{\rho_2}{T_2^{\downarrow}} + K \epsilon \rho_3$$

$$(6c) \quad \frac{\partial \bar{\rho}_3}{\partial t} = -\frac{\rho_3 - \rho_3}{T_1} - K \epsilon \rho_3$$

Where we consider T_1 as inversion decay time, $T_2^{\downarrow}$ as dipole moment decay time and $\bar{\rho}_3^{\downarrow}$ as the value of inversion in absence of the external field ($\epsilon=0$). Equation (6c) gives us information about the rate of variation of the inversion i.e. the population density at the excited level. On the other hand solution² of equation (2) as proposed by Rabi leads to an equation giving the transition probability as a function of time:

$$(7a) \quad |C_a(t)|^2 = \left[\frac{\sin^2(\Omega t/2)}{(\Omega/2)^2} \right] \left(\frac{\kappa \epsilon_0}{2} \right)^2$$

In fact taking care of various decaying factors present in the medium, equation (7a) can be written as

$$(7b) \quad |C_a(t)|^2 = \frac{1}{4} \exp(-\gamma_b t) \left[\frac{\sin^2(\Omega t/2)}{(\Omega/2)^2} \right] \left(\frac{\kappa \epsilon_0}{2} \right)^2$$

The above equation indicates a periodic variation in the transition probability of the dipoles, which arises out of nutation of the dipole vectors in presence of the laser field at a frequency given by

$$(7c) \quad \Omega(\Delta) = \sqrt{\Delta^2 + (\kappa \epsilon_0)^2}$$

Where $\Delta = \omega_0 - \omega$ is the detuning frequency (i.e. the frequency difference between the laser field and the dipole transition frequency), ϵ_0 is the steady value of the applied field, κ is defined in terms of the dipole moment of the molecules as

$$(8) \quad \kappa = \frac{2d}{\hbar}$$

Equations (7b) and (7c) are the keys to our explanation of the experimental result. Using equation (7b) we have sketched graphs showing the time evolution of population in the upper state (inversion) for five different values of detuning at 100, 110, 120, 130 and 140 MHz.

An important feature of the graphs can be seen from fig.2 is that all the graphs come to a common phase representing minimum value of transition probability $|C_a(t)|^2$ at an interval 6^2n ($n=, 1, 2, \dots$ e.t.c.). Again as predicted by equation (7c), the frequency of nutation i.e. the periodic variation of transition probability is a function of detuning. This feature is clearly present in fig.2 also. Because in the interval 0 to 6^2n as the detuning varies from 100 to 110, the transition frequency changes from 10 to 11. In other words for $\partial\Delta \rightarrow 10$ units; $\delta\Omega \rightarrow 1$ unit.

Vector representation of transition probability: The maxima of the transition probabilities corresponding to different values of detuning are joined by straight arrows representing vectors.

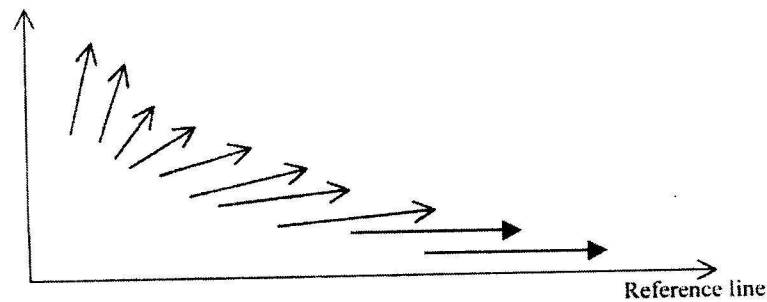
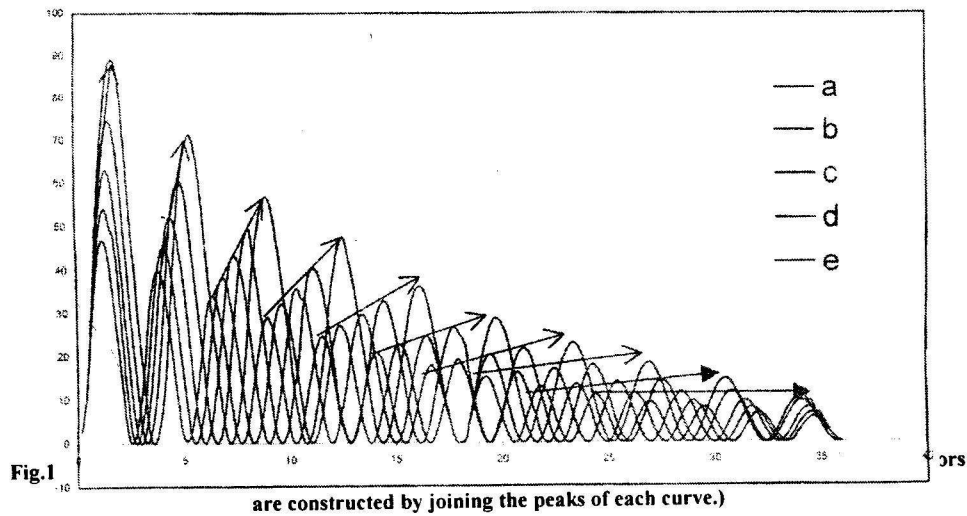


Fig.2 (Vectors representation of transition probability)

The periodic fluctuation of transition probability in presence of laser can be interpreted by these vectors representation as follows: By considering the x-axis as the reference line it can be observed that the orientation of these representative vectors towards the reference line indicates total dephasing of the transition probabilities of different groups under consideration and on the other hand the orientation of the vectors at right angle to the reference line represents in-phase condition of the transition probabilities for all the concerned groups fig.2. This vector representation can be a very handy tool in understanding the behaviour of a medium in presence of a strong resonant field. The interesting point to note in the graphical representation is the periodic accumulation of the transition probabilities to a common phase after a certain time interval, which of course is caused by beats formation between two periodic functions with slight difference in frequency.

3. Experiment

A π -type discharge tube of length 20cm having construction in the middle has been used to obtain a continuous discharge with a typical white colour representing the Angstrom band of CO appearing in the +ve column. A constant low pressure was ensured between the electrodes of the tube. The potential difference between the electrodes is adjusted just above threshold and a smooth discharge appears in the tube. Now as the 500mw laser is switched on and focussed at the +ve column of the discharge, the current drops down to almost half its initial value for a very short period of time and then shows a very small amount of periodic fluctuation as shown in table-1.

Table-I

Sl. No.	Gas pressure	Threshold voltage	Discharge current (without laser)	Discharge current (with laser) Current in ma.		Remark
				Time		
1	1 torr.	2000V	8.1 ma.	1 sec	7.0 ma	As the laser is switched off the discharge current rises to its former level again..
2	"	2000V	"	5 sec	7.0 ma	
3	"	2000V	"	10 sec	7.1 ma	
4	"	2000V	"	15 sec	7.1 ma	
5	"	2000V	"	20 sec	7.0 ma	
6	"	2000V	"	25 sec	7.1 ma	
7	"	2000V	"	30 sec	7.1 ma	
8	"	2000V	"	35 sec	7.1 ma	

4. Result and discussion

The discharge current that was steady under normal laboratory condition showed a sudden dip as the discharge was exposed to the laser radiation. This dip in the discharge current due to application of the Ar^+ laser can be explained as follows;

Molecules of the medium under consideration have different amount of detuning with the laser field because of their thermal motion (Doppler broadening). As their thermal velocity distribution follows classical Maxwell Boltzmann law, the detuning can vary continuously in a particular range for a certain thermodynamical state of the medium. Let us divide the whole medium into N groups of molecules with a small range of detuning $\partial\Delta_1, \partial\Delta_2, \partial\Delta_3, \dots, \partial\Delta_N$. As given by equation (7c), these groups will undergo nutation at frequencies $\Omega_1, \Omega_2, \Omega_3, \dots, \Omega_N$ respectively. This will bring all the groups out of phase with each other so that if for one group the transition probability $|C_a(t)|^2$ is maximum, for the next group it is away from maximum and so on. However as two periodic functions with slight frequency difference produce beats, these groups also do the same and thereby out of these N groups N/2 evolve that will undergo nutation with frequencies

$\Omega' = \Omega_1 - \Omega_2, \Omega'' = \Omega_3 - \Omega_4, \dots$ etc. These $N/2$ periodic functions also will produce beats and thereby evolve $N/4$ number of groups with periodicity of nutation, which is much smaller than the initial frequency with which the nutation started. This process goes on until the beat frequency or the nutation frequency $\Omega \rightarrow 0$. At this stage the nutation is said to have died down.

It should be noted that the nutation frequency at the beginning is of the order of GHz and so it is too high to be observed in our experimental set up. However as a result of continuous beating among the respective groups we will arrive at a situation where the beat frequency (i.e. the frequency of the periodic fluctuation of transition probability) is low enough to be observed by our detecting meters.

5. Conclusion

We attribute this dip in discharge current to the nutation of the gas molecules inside the tube. The dephasing of the molecular groups caused by nutation is broken at a certain interval when all the groups find themselves at lowest value of the transition probability. This is the point when the discharge current dips. This particular situation can be best analysed and understood with the help of vector representation. Finally it can be noted that the continuous beating among the groups undergoing nutation reduces the nutation frequency to a range so that the detecting devices used in the present experiment can observe it.

Acknowledgement

The authors are thankful to Prof. D.K. Rai, BHU, Varanasi for his helpful discussion.

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