METASTABLE QUENCHING BY OPTICAL PUMPING

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Abstract

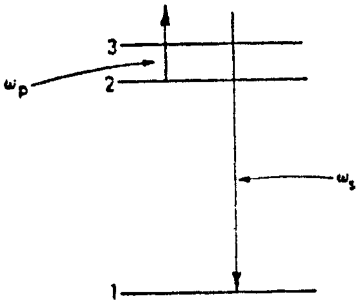
A theoretical study of the quenching of the metastable states in hydrogen by optical coupling to energetically close radiative states is presented. The dependence of the metastable lifetime on the intensity and frequency of the optical radiation is determined. The advantages of using an optical field over static electric field are described. Applications to optically controlled switches, and in particular, the effect of controlling metastable populations are discussed.

Introduction

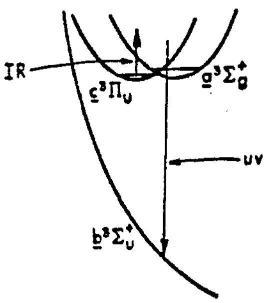
Metastable states in atoms or molecules play an important role in the dynamics of neutral and ionized gases and beams. In electric discharges they provide an intermediate state in the step ionization of neutral species. The metastable states constitute a reservoir of energy, and are therefore important in laser plasmas. They are also of interest in connection with the problem of forming energetic neutral molecular beams.

Many optical processes may be considered for gas phase switch control affecting the initiation, conduction, and recovery periods.[1-3] For example, in applications of the optogalvanic effect, the discharge properties are controlled by illuminating the discharge with (laser) light. As an example of this, removal of the metastable species in neon discharges by laser radiation has been studied recently.[4] Quenching of the metastable states in hydrogen molecules by the application of a static electric field has also been studied.[5,6] This work reports a study of the metastable quenching in a molecular hydrogen three level system by the application of either a static or optical electric field. A short summary only is presented here; detailed results are to be published.[7]

Theory



1(a)



1(b)

Fig. 1a. Energy level diagram for the three level system. ω_p and ω_s denote the frequencies of the pump and the signal fields

Fig. 1b. Potential energy curves (not to scale) for the three lowest triplet states in H_2.

The interaction of a homogeneously broadened three level atomic or molecular system with two electric fields is studied using the semiclassical density matrix formalism. The energy level diagram for the system is shown in Fig. 1. The transition between state 1 and 2 assumed to be forbidden, so that the dipole moment $\mu_{12}=0$. Figure 1b shows the potential energy curves for the three lowest states in hydrogen. Since off-resonant optical pumping as well as pumping by a static field are considered here, the frequency mismatches between the applied fields and the transitions are kept arbitrary. The intensities of the two fields and the lifetimes of the three levels involved are also kept arbitrary in this study. The density matrix equations for the system can be written as:

$$\dot{\rho}_{11} = \frac{\rho^e_{11} - \rho_{11}}{T_1} + \frac{\mu_{13}E}{i\hbar}(\rho_{13} - \rho_{31})$$

$$\dot{\rho}_{22} = \frac{\rho^e_{22} - \rho_{22}}{T_2} + \frac{\mu_{23}E}{i\hbar}(\rho_{23} - \rho_{32})$$

$$\dot{\rho}_{33} = \frac{\rho^e_{33} - \rho_{33}}{T_3} - \frac{\mu_{13}E}{i\hbar}(\rho_{13} - \rho_{31}) - \frac{\mu_{23}E}{i\hbar}(\rho_{23} - \rho_{32}) \tag{1}$$

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$$\dot{\rho}_{13} = -(i\omega_{13} + T_{13}^{-1}) \rho_{13} + \frac{\mu_{13}E}{i\hbar} (\rho_{11} - \rho_{33}) + \frac{\mu_{23}E}{i\hbar} \rho_{12}$$

$$\dot{\rho}_{12} = -(i\omega_{12} + T_{12}^{-1}) \rho_{12} + \frac{\mu_{23}E}{i\hbar} (\rho_{13}) - \frac{\mu_{13}E}{i\hbar} \rho_{32}$$

$$\dot{\rho}_{32} = -(i\omega_{32} + T_{32}^{-1}) \rho_{32} + \frac{\mu_{23}E}{i\hbar} (\rho_{33} - \rho_{22}) - \frac{\mu_{13}E}{i\hbar} \rho_{12} \qquad (2)$$

$$E = \frac{E_p}{2} e^{i\omega_p t} + \frac{E_s}{2} e^{i\omega_s t} + \text{complex conjugate}$$

Here ρ_{ii}^e denote the values of the density matrix elements ρ_{ii} in the absence of the fields. T_1, T_2, T_3 denote the natural lifetimes for the three levels. We assume $T_{ij}^{-1} = 1/2(T_i^{-1} + T_j^{-1})$.

The density matrix equations have been solved previously in the steady state[8] assuming $T_1 = T_2 = T_3$ and in the time dependent case[9] ignoring the natural decay. To determine the effect of electric field quenching, the transient solutions for the general case of $T_1 \neq T_2 \neq T_3$ are needed. The rotating wave approximation, appropriate for the cases of optical pumping and static pump field is invoked to simplify equations 1 and 2. The simplified equations are then solved in the transient situation by first converting them into a set of algebraic equations by Laplace transformation assuming that during the pumping period the field amplitudes remain constant. Moreover we assume that the pumping starts at t=0 and that the initial conditions are $\rho_{11}(0) = \rho_{33}(0) = 0$, $\rho_{ij}(0) = 0$ for $i \neq j$ and $\rho_{22}(0) = 1$. This means that all molecules are in state 2 at t=0. Also, it is assumed that the level populations are changing only by electric field pumping and the natural decay, so that $\rho_{ii}^e = 0$. The algebraic equations obtained by Laplace transformations are solved. The Laplace inverse transforms of the solutions then provide the time dependent values of the density matrix elements.

Results

In this section some applications of the theory are summarized. The case of the H_2 triplet system is considered. The level 1 is the dissociative $\underline{b}\,^3\Sigma_u^+$ state for which $T_1 = 10^{-14}$ s. Levels 2 and 3 correspond to the metastable $\underline{c}\,^3\Pi_u$ and the radiative $\underline{a}\,^3\Sigma_g^+$ states respectively so that in absence of collisional quenching, $T_2 = 10^{-3}$ s and $T_3 = 10^{-8}$ s. If the effect of collisions is included, we have

$$T_2 = (6.65 \times 10^6\ P + 10^3)^{-1}\ s$$

$$T_3 = (6.65 \times 10^6\ P + 10^8)^{-1}\ s$$

where P is the pressure of the gas in Torr. Here it is assumed that all the collisions are 'hard.'[7] The dipole moments for both the transitions are assumed to be 1 Debye.

From the time dependence of the metastable population ρ_{22}, the e^{-1} lifetime T_2 is determined. In Figures 2 and 3 T_2 is plotted as a function of the pump field intensity. In Figure 2 the effect of collisions are included. In both figures, both the small signal and the strong signal cases are shown. The off-resonance case shown corresponds to pumping by a CO_2 laser line. We note that appreciable quenching of the metastable state by an off-resonant pump requires very large pump intensities. For a pump beam on resonance, the pump intensity required is several orders of magnitude smaller. Although a specific case has been considered here, application of the theory developed to other systems such as Hg_2 or the noble gases, is straightforward.

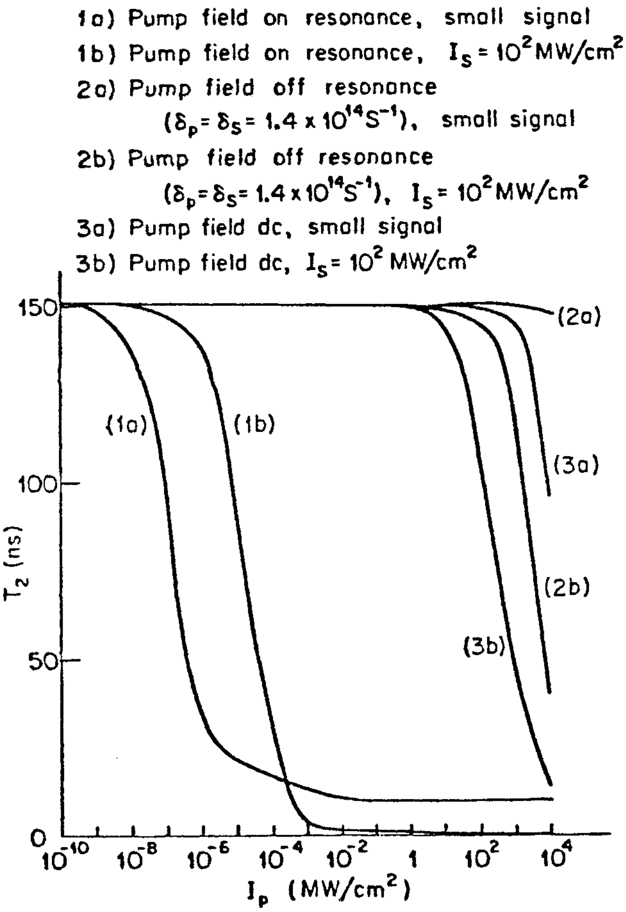

Fig. 2. The e^{-1} lifetime of the metastable $\underline{c}\,^3\Pi_u$ state in H_2, as a function of the pump intensity I_p. A gas pressure of 1 Torr is assumed.

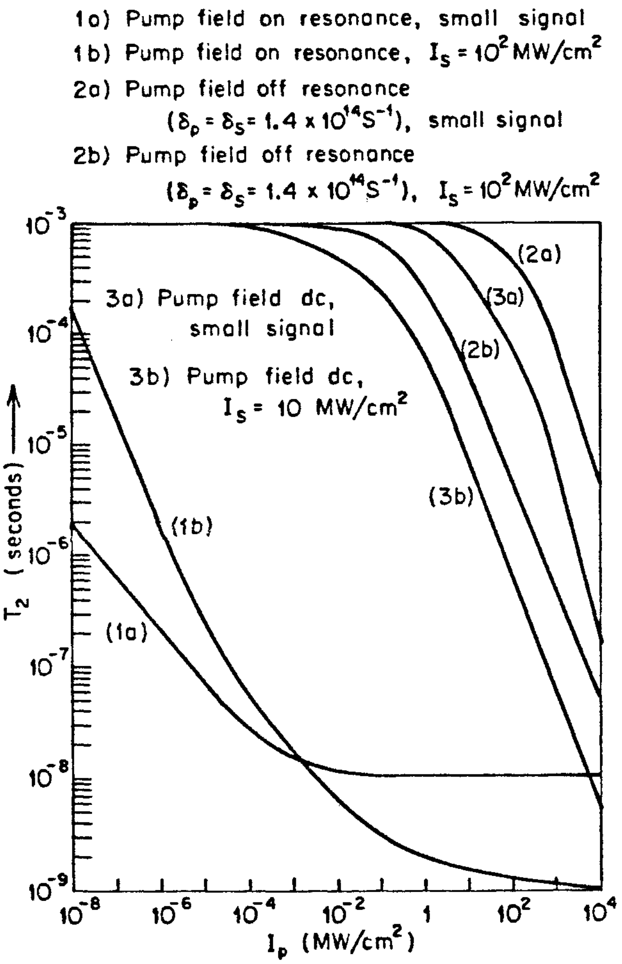

Fig. 3. The e^{-1} lifetime of the metastable $c^3\Pi u$ state in H_2 as a function of the pump intensity I_p, when the quenching by collisions is ignored.

Summary

The quenching of metastable states by the application of electric fields is studied theoretically. Numerical results are presented for the application to a specific system (the hydrogen molecular triplet). The effect of signal power saturation on the metastable lifetime is shown. The theory consists of an extension of the density matrix formalism to the cases of 1) unequal natural lifetimes of the levels, 2) transient situation, and 3) static pump field.

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