Formation and stimulated photodissociation of metastable molecules with emission of photon at the collision of two atoms in a laser radiation field

E Gazazyan and A Gazazyan

Institute for Physical Research, NAS of Armenia, Ashtarak-2, 0203, Armenia

E-mail: emilgazazyan@gmail.com

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Abstract
The formation of metastable molecules (Feshbach resonances) at the collision of two atoms and subsequent stimulated transition to a lower unbound electronic molecular state, with emission of a photon of the laser radiation has been investigated. This can develop, in particular, for $\text{Rb}_2$ molecules due to resonance scattering of two $\text{Rb}$ atoms. This process is a basis for the creation of excimer lasers. Expressions have been obtained for the cross sections of elastic and inelastic resonance scattering and the intensity of the stimulated emission of the photons.

Keywords: Feshbach resonances, metastable molecules, excimer laser

(Some figures may appear in colour only in the online journal)
fields of physics. It is e.g. used for explanation of asymmetry in the absorption impurity ions in crystals, which is caused by formation of excitonic resonances [16, 17]. With use of these resonances works [18, 19], study the phenomenon of storage and reconstruction of quantum information in solids. The asymmetric form of the Fano resonance is important in considerations of nanoscale structures of interacting quantum systems [20, 21].

In the present work we consider collision of atoms with formation of metastable molecules (Feshbach resonances) and the subsequent stimulated transition to the lower unbound molecular electronic states with the emission of a photon of laser radiation. Expressions for the cross sections of the elastic and inelastic resonance scattering and the intensity of the stimulated emission of the photons have been obtained.

2. Formation of Feshbach resonance

Consider elastic and inelastic collision of two atoms with formation of Feshbach resonance (figure 1). Let $U$ be the interaction which couples electronic states in the open and closed channel. Laser radiation with frequency $\omega$ couples the upper molecular quasi bound state with the lower uncoupled molecular state with interaction $\Omega$. The Hamiltonian for the considered process (figure 1) has the following form:

$$H = E_e|e\rangle\langle e| + \int dE E\left(|E\rangle_1\langle E| + |E - \omega\rangle_2\langle E - \omega|\right)$$

$$+ \int dE U|e\rangle_1\langle E| + U'|E\rangle\langle e'|$$

$$+ \int dE \left(\Omega_{E-\omega}(t)|e\rangle_2\langle E - \omega| + \Omega_{E-\omega}'(t)|E - \omega\rangle_2\langle e'|\right).$$

(1)

We represent the solution of the Schrödinger equation with the Hamiltonian (1) as:

$$|\Psi(t)\rangle = C_1(t)|e\rangle e^{-iEt} + \int dE e^{-iEt}b_{1,E}(t)|E\rangle_1$$

$$+ e^{iEt}b_{2,E-\omega}(t)|E - \omega\rangle_2.)$$

(2)

Substituting expression (2) for the wave vector and using that $\Omega = \Omega e^{-i\delta}$, where $\omega$ is the frequency of laser radiation, into the Schrödinger equation we obtain the following system of differential equations for the coefficients of the expansion of (2):

$$i\frac{dC_1(t)}{dt} = \int dE \left(U|e\rangle_1\langle E| + U'|E\rangle\langle e'|\right)$$

$$+ \int dE \left(\Omega_{E-\omega}(t)|e\rangle_2\langle E - \omega| + \Omega_{E-\omega}'(t)|E - \omega\rangle_2\langle e'|\right).$$

(3)

After the Fourier transform for the coefficient in the formula (2) we obtain for the Fourier components of the expansion coefficient the following system of equations:

$$\lambda - E_1 C_1(\lambda) = \int dE b_{1,E} b_{1,E}(\lambda)$$

$$b_{1,E}(t) = \int d\lambda e^{-i\lambda E}C_1(\lambda)$$

$$b_{2,E-\omega}(t) = \int d\lambda e^{-i\lambda E - i\omega}\Omega_{E-\omega}(\lambda)$$

(4)

Now we obtain from the equations (6) and (7) [10]:

$$b_{1,E}(\lambda) = \frac{P_1}{\lambda - E + Z(\lambda)\delta(\lambda - E)}$$

$$b_{2,E-\omega}(\lambda) = \Omega_{E-\omega}(\lambda)$$

(8)
\[ b_{2,E-\omega}(\lambda) = \left[ \frac{P}{\lambda - E} + Z(\lambda)\delta(\lambda - E) \right] \Omega_{E-\omega}^\dagger C(\lambda), \] (9)

where
\[ Z(\lambda) = \frac{\lambda - E_\omega - \Delta(\lambda)}{\Gamma(\lambda)}. \] (10)

In expression (10) \( \Delta(\lambda) \) and \( \Gamma(\lambda) \) are the full resonant shifts and width and \( P \) denotes the principle value:
\[ \Delta(\lambda) = \Delta_r(\lambda) + \Delta_i(\lambda) \]
\[ \Delta_r(\lambda) = P \int dE \frac{|U_E|^2}{\lambda - E} \]
\[ \Delta_i(\lambda) = P \int dE \frac{|\Omega_{E-\omega}|^2}{\lambda - E} \]
\[ \Gamma(\lambda) = \Gamma_r(\lambda) + \Gamma_i(\lambda) \]
\[ \Gamma_r(\lambda) = 2\pi|U_E|^2 \]
\[ \Gamma_i(\lambda) = 2\pi|\Omega_{E-\omega}|^2. \]

By substituting expressions (8) and (9) into formula (2) from the orthonormalization condition we obtain the following expression for the \( C(\lambda) \):
\[ C(\lambda) = \sqrt{\frac{2\pi}{\Gamma(\lambda)z^2(\lambda) + \pi^2}}. \] (11)

For the first solution we obtain the following expression:
\[ |\Phi_\lambda^{(1)}(t)| = c_\lambda(e)\left[ |e\rangle + \int dE \left( \frac{P}{\lambda - E} + z(\lambda)\delta(\lambda - E) \right) \right] \]
\[ e \left( U_E^\dagger|E\rangle_1 + e^{i\omega\int|\Omega_{E-\omega}|^2|E - \omega\rangle_2} \right). \] (12)

For the second orthonormalized solution in the case of \( c_\lambda(\lambda) = 0 \), we have the following expression:
\[ |\Phi_\lambda^{(2)}(t)| = \sqrt{\frac{2\pi}{\Gamma(\lambda)}} \left( \Omega_{E-\omega}|\lambda\rangle_1 - e^{i\omega\int|U_{E}|^2|\lambda - \omega\rangle_2} \right). \] (13)

These solutions (12) and (13) provide the orthonormalization condition for quasienergy functions:
\[ \langle \Phi_\lambda^{(1)}(t)|\Phi_\lambda^{(2)}(t)\rangle = \delta_{\lambda,\lambda'}\delta(\lambda' - \lambda). \] (14)

3. Cross sections of elastic and inelastic scattering and intensity of emission radiation

Asymptotic \((r \to \infty)\) expressions for continuous-spectrum wave functions with orbital angular momentum \( l \) are known to have the following appearance:
\[ |E_l^{(1)}(E)| \propto \frac{1}{k_{r1}} \sin \left( k_{r1}r + \delta_1 - \frac{1}{2}k_{r1} \right) P_l(\cos \Theta_1), \]
\[ k_1 = k(E) \] (15)
\[ |E - \omega_l^{(1)}(E)| \propto \frac{1}{k_{r2}} \sin \left( k_{r2}r + \delta_2 - \frac{1}{2}k_{r2} \right) P_l(\cos \Theta_2), \]
\[ k_2 = k(E - \omega) \] (16)

with \( P(\cos \Theta) \) being the Legendre polynomials. Taking into account that the wave functions of bound states of atoms vanish asymptotically \((|e\rangle = 0)\) at large distance \((r \to \infty)\) we can write the quasienergy wave functions (12) and (13) as follows:
\[ |\Phi_\lambda^{(1)}(t)| = -\frac{\pi c(\lambda)}{\sin \eta} \left( U_{E1}^{\dagger} \sin \left( k(\lambda)r_1 + \eta + \delta_1 - \frac{\eta}{2} \right) P_\eta(\cos \Theta_1) \right) \]
\[ + e^{i\omega\int|\Omega_{E-\omega}|^2|k(\lambda - \omega)r_2 + \eta + \delta_2 - \frac{\eta}{2}|| P_\eta(\cos \Theta_2) \right) \] (17)
\[ |\Phi_\lambda^{(2)}(t)| = \sqrt{\frac{2\pi}{\Gamma(\lambda)}} \left( \Omega_{E-\omega}|\lambda\rangle_1 - e^{i\omega\int|U_{E}|^2|\lambda - \omega\rangle_2} \right) \]
\[ P_\eta(\cos \Theta_1) \]
\[ - e^{i\omega\int|U_{E}|^2|\lambda - \omega\rangle_2} \left( \sin \left( k(\lambda - \omega)r_2 + \eta + \delta_2 - \frac{\eta}{2} \right) P_\eta(\cos \Theta_2) \right) \] (18)

where \( \delta_l \) is the phase of non-resonant scattering and \( \eta \) is the phase caused by resonant Feshbach scattering
\[ \tan \eta = -\frac{\pi}{z(\lambda)}. \] (19)

We now represent the scattering state vector \( |\Phi_\lambda(1 \to 1, 2)\rangle \) at \( r \to \infty \) as superposition of quasienergy function (17) and (18)
\[ |\Phi_\lambda(1 \to 1, 2)\rangle = \sum_j A_j|\Phi_\lambda^{(j)}(t)| \] (20)

with
\[ \sum_j |A_j|^2 = 1 \] (21)
then, if we require the presence of incoming and outgoing waves in the first, elastic, channel and the absence of any incoming wave in the second, inelastic, channel we can write for the expansion coefficients in (20) $A_j$ the following:

$$A_1 = \frac{U_\lambda}{\sqrt{|U_\lambda|^2 + |\Omega_\lambda|^2}} , \quad A_2 = \frac{\Omega_\lambda e^{-i\eta}}{\sqrt{|U_\lambda|^2 + |\Omega_\lambda|^2}} .$$  \hspace{1cm} (22)

From expressions (22) we can obtain for the scattering state vector (20):

$$|\Phi_1(1, 2)\rangle = \left[ \frac{e^{i\lambda r_2}}{k(\lambda) r_2} \right] \sin(k(\lambda)r_1 - \frac{i\eta}{2}) - \frac{e^{i\eta}}{2r_2} \frac{\Gamma_\lambda(\lambda)}{\Gamma(\lambda)} \left( 1 - e^{2i\eta} \right)$$

$$+ e^{-2i\eta} \left[ \frac{e^{i\lambda r_2}}{k(\lambda) r_2} \right] \cos(\Theta_2) - e^{i\omega_2} \frac{\sin \eta}{k(\lambda - \omega)r_2} \sqrt{\frac{\Gamma_\lambda(\lambda)}{\Gamma(\lambda)}}$$

$$\times e^{i\omega_2} e^{-i\lambda r_2} - \frac{e^{i\eta}}{2r_2} P_{21}(\cos \Theta_2).$$  \hspace{1cm} (23)

From expression (23) we can obtain the formulas for corresponding cross sections

$$\sigma(1 \rightarrow 1) = \frac{4\pi(2l + 1)}{k^2(\lambda)} \left[ \frac{\Gamma_\lambda(\lambda)}{\Gamma(\lambda)} \right] \left( 1 - e^{2i\eta} \right) + e^{-2i\eta} - 1 \right|^2$$

$$\sigma(1 \rightarrow 2) = \frac{4\pi(2 + 1)}{k^2(\lambda - \omega)} \frac{\Gamma_\lambda(\lambda)\Gamma_\lambda(\lambda)}{\Gamma(\lambda - \omega)} \sin^2 \eta.$$  \hspace{1cm} (24)

Let us separate in (24) for elastic scattering the resonant state and write the potential part of scattering cross sections in the form

$$\sigma_{pot} = \frac{\pi(2l + 1)}{k^2(\lambda)} 4 \sin^2 \delta_i.$$  \hspace{1cm} (26)

The total cross section of elastic scattering we write as:

$$\sigma_{tot}^el = \sigma_{pot} + \sigma_{res}^el$$  \hspace{1cm} (27)

where

$$\sigma_{res}^el = \frac{\pi(2l + 1)}{k^2(\lambda)} \frac{\Gamma_\lambda^2(\lambda)}{\Gamma(\lambda)} |1 - e^{2i\eta}|^2.$$  \hspace{1cm} (28)

From expressions (10) and (19) for the resonant elastic and inelastic cross sections, we obtain:

$$\sigma_{res}^el = \frac{\pi(2l + 1)}{4k^2(\lambda)} \frac{\Gamma_\lambda^2(\lambda)}{\lambda - E_\epsilon - \Delta(\lambda)^2 + \frac{\Gamma(\lambda)}{4}}$$

$$\sigma_{res}^{inel} = \frac{\pi(2l + 1)}{k^2(\lambda)} \frac{\Gamma_\lambda(\lambda)\Gamma_\lambda(\lambda)}{\lambda + \omega - E_\epsilon - \Delta(\lambda)^2 + \frac{\Gamma(\lambda)}{4}} .$$  \hspace{1cm} (29)

Distribution for the spectral intensity of the emission radiation at the stimulated transition to the lower unbound molecular state has the following form [22]:

$$I(\omega) = 4\pi I_0 \frac{1}{\omega - E_\epsilon - \Delta(\lambda)^2 + \frac{\Gamma(\lambda)}{4}} .$$  \hspace{1cm} (30)

where $I_0$ is the full intensity of the incident laser radiation field then for the intensity of the stimulated emission of photons coming from unit volume of the gas are obtained:

$$I(\omega) = N^4 I_0 \frac{1}{2\pi} \left( \omega + \lambda - E_\epsilon - \Delta(\lambda)^2 + \frac{\Gamma(\lambda)}{4} \right)^2 .$$  \hspace{1cm} (31)

The spectral intensity of the stimulated emission of photons coming from unit volume of the gas are obtained:

$$I(\omega) = N^4 I_0 \frac{1}{2\pi} \left( \omega + \lambda - E_\epsilon - \Delta(\lambda)^2 + \frac{\Gamma(\lambda)}{4} \right)^2 .$$  \hspace{1cm} (32)

Figures 2–4, show the curves corresponding to the formulas ((29), (30) and (32)), for the values $\Gamma_\lambda = 10^\nu$ Hz and $\Gamma_\lambda = 10\Gamma_\lambda$, where $N^4$ is the density of excited atoms in the gas. It is seen from expression (32) that at high concentrations of excited atoms in the gas we have an intense stimulated radiation.

It should be noted that in the case of dense gases consideration should be given to cooperative effects in the collision of atoms. We will consider these phenomena in future studies.

4. Conclusion

We consider collision of two atoms with the formation of metastable molecules (Feshbach resonance) and the subsequent stimulated transition under the influence of a laser radiation field to a lower unbound molecular electronic state with emission of a photon of laser radiation. This situation can develop, in particular, for $Rb_2$ molecules due to resonance scattering of two $Rb$ atoms in states $5s$ and $5p$, with the formation of metastable molecular state $^3\Pi_u$, with subsequent stimulated transition to the lower $^1\Sigma_u^+$ unbound molecular electronic state, with the emission of photons of laser radiation. Expressions for the cross sections of the elastic and inelastic resonance scattering and the intensity of the stimulated emission of photons coming from unit volume of the gas are obtained.

The typical atomic gas density is $10^{14}$–$10^{17}$ cm$^{-3}$. If the density of excited atoms is 0.001%, then the intensity of emitted photons during the simulated transition is $10^9$–$10^{12}$ times larger. As a result, we have a source of high radiation, which serves as an example of an excimer laser. From the expression (32), the peak of the spectral intensity in the graph at $\omega = E\epsilon + \Delta(\lambda) - \lambda$ is $I(\omega) = N^4 I_0^{2/\pi} .$

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