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RADIATIVE TRANSITIONS IN ATOM-ATOM SCATTERING IN INTENSE LASER FIELDS*

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ABSTRACT

The quantum nonperturbative analysis of Kroll and Watson for a 2-level system of near-adiabatic atom-atom scattering in an intense laser field mode is extended to treat a general multilevel system interacting with intense single or many field modes. A procedure for solving rigorously the adiabatic eigenvalue problem for the whole charge-field system is given. A new transition probability formula is derived. Cross sections are calculated for the processes $Li + H(X^1\Sigma^+) + \nu\hbar\omega \rightarrow Li + H(A^1\Sigma^+$ or $B^1\Pi)$ where $\nu \geqslant 1$. Analysis of transition in an atom due to intensity variation of a laser pulse shows that desired transition probability (e.g., 1/2) per pulse may be achieved by varying the pulse parameters. For this, numerical results of $Na(3s) + 2\hbar\omega \rightarrow Na(5s)$ and of $Li(2s) + 8\hbar\omega \rightarrow Li(3s)$ are given.

I. INTRODUCTION

In this paper, we analyze two physical situations: (i) near-adiabatic atom-atom[1] collision in an intense laser beam; and (ii) an atom[1] being irradiated by a strong laser pulse. We are concerned with the calculation of probability of single/multiphoton bound-bound transition in the atomic system. There have been many experiments on multiphoton transitions in atoms and molecules in intense laser field.[2-5] Resonant bound-bound transitions are often decisive in multiphoton ionization results.[2,4] Most theoretical analyses on this subject deal with isolated atoms or molecules.[6-9] There are comparatively fewer theoretical studies of atom-atom collision in an intense laser field.[10,11] In experiment with an atomic gas, radiative transition during atom-atom collision is significant compared to that of the isolated atoms.[5]

Kroll and Watson[10] (hereafter referred to as KW) have analyzed the interaction of a two-level "quasi-molecule" (the atom-atom in near-adiabatic collision) with an intense radiation mode. With similar approach, the present paper analyzes more general cases of a quasi-molecule of finite number ($n \geqslant 2$) of discrete levels interacting with a finite number of intense field modes, thus providing treatment for greater variety of physical phenomena. Approximating the real quasi-molecule with more-than-two levels is useful since the role of near-resonant intermediate states in determining the multiphoton transition probability and of nonresonant levels in determining the energy shift are important. The analysis does not make any perturbation expansion nor the rotating wave approximation.[9] It is not limited to the electric dipole approximation.

In Section II, we write down the time-dependent equation for near-adiabatic atom-atom collision in intense laser modes. Level shifts and coupling between states are found from solutions of the adiabatic molecular eigenvalue problem in Sec. III. In Sec. IV, a new formula of transition probability between two shifted levels is derived. Section V contains two numerical studies. Finally in Sec. VI, we study transition in an isolated atom due to intensity variation of the irradiating laser pulse.

II. TIME-DEPENDENT EQUATION

We consider near-adiabatic scattering of atoms in m field modes in a large cavity. The eigenvalues and eigenfunctions of the adiabatic molecular hamiltonian h are written as w_α and ϕ_α respectively, with parametric dependency on the fixed internuclear configuration $\underset{\sim}{R}$.[12] Let the free-field hamiltonian and the charge-field interaction hamiltonian be h_γ and h' respectively.[13] We shall approximate the relative motion of the nuclei by classical orbits $\underset{\sim}{R}(t)$. Then in the c.m. frame of the quasi-molecule, the hamiltonian describing the whole charge-field system is $H_c(t) = h + h_\gamma + h'$, where h and h' are functions of the orbit $\underset{\sim}{R}(t)$.[10]

Since in scattering experiments, the initial state of the quasi-molecule is prepared before the charge-field interaction takes place and the final state is observed after the interaction has occurred, we therefore expand the total wavefunction

$$\psi = \sum_{\beta=1}^{n} c_\beta(t)\,\phi_\beta \,.$$

Substitution into the time-dependent Schrödinger equation for $H_c(t)$ gives

$$i\hbar\dot{c}_\alpha = w_\alpha c_\alpha + h_\gamma c_\alpha + \sum_{\beta\neq\alpha} (\phi_\alpha, h'\phi_\beta) c_\beta \,, \tag{2.1}$$

where the term $(\phi_\alpha, h'\phi_\alpha)$ for radiative transition between nuclear molecular states of the same electronic state and the term $i\hbar \sum_\beta c_\beta(\phi_\alpha, \dot{\phi}_\beta)$ for collisional transition have been neglected.[10]

We now expand the c_α in terms of the photon number states $\Omega(N_\lambda - \nu_\lambda)$ where N_λ is the initial mean number of photons in the λth mode and $\nu_\lambda > 0$ (<0) is the number of photons absorbed (emitted) by the quasi-molecule. That is

$$c_\alpha = \sum_{\{\nu_\lambda\}} i^{\sum_\lambda \nu_\lambda}\, b_{\{\nu_\lambda\}}(\alpha)\, e^{-\frac{i}{\hbar}\int^t (w_p + \sum_\lambda \hbar\omega_\lambda N_\lambda)dt'} \prod_\lambda \Omega(N_\lambda - \nu_\lambda) \,, \tag{2.2}$$

where w_p is a particular w_α chosen for convenience of calculation, and $\{\nu_\lambda\}$ denotes a set of m integers, corresponding to m modes. Thus $b_{\{\nu_\lambda\}}(\alpha)$ is the probability amplitude that the charge-field system is in the electronic state α with $\{\nu_\lambda\}$ photons "absorbed". With the excellent approximation for intense field modes

$$N_\lambda - \nu_\lambda + 1 \approx N_\lambda - \nu_\lambda \approx N$$

we obtain from Eqs. (2.1) and (2.2)

$$i\frac{d}{dy} b_{\{\nu_\lambda\}}(\alpha) = W_{\{\nu_\lambda\}}(\alpha)\, b_{\{\nu_\lambda\}}(\alpha) + \sum_{\gamma=1}^{m} \sum_\beta G_\gamma^{\pm}(\alpha,\beta)\Big(b_{\{\nu_\gamma-1\}}(\beta) \pm b_{\{\nu_\gamma+1\}}(\beta)\Big) \,, \tag{2.3}$$

where $y \equiv vt/a_o$ is a dimensionless time variable defined in terms of any convenient constant speed v and Bohr radius a_o; and

$$W_{\{\nu_\lambda\}}(\alpha) \equiv \frac{a_o}{\hbar v}(w_\alpha - w_p) - \sum_\lambda \nu_\lambda F_\lambda$$

is the "unperturbed eigenlevels" of the noninteracting hamiltonian $h + h_\gamma$; and

$$F_\lambda \equiv a_o\omega_\lambda/v$$

is the "photon energy". In the subscript set $\{\nu_\gamma \pm 1\}$ of the last term in Eqs. (2.3), all the component indices are the same as those in the set $\{\nu_\lambda\}$ of the first and second terms except the γth, for which one has $\nu_\gamma \pm 1$ instead. The upper and lower sign in Eqs. (2.3) correspond to the use of the electric dipole[14] interaction hamiltonians,[15]

$$h'_+ \equiv -\sum_i q_i \mathbf{r}_i \cdot \mathbf{E}(0)$$

and

$$h'_- \equiv -\sum_i \frac{q_i}{m_i c} \mathbf{p}_i \cdot \mathbf{A}(0)$$

respectively. We have used both forms in our numerical calculations, though it is believed that the h'_+ is a better approximation in treatment where higher levels are neglected.[16] For plane wave modes,

$$G^\pm_\lambda(\alpha,\beta \neq \alpha) \equiv \frac{a_o}{\hbar v}\left(\frac{2\pi I_\lambda}{c}\right)^{\frac{1}{2}}\left(\phi_\alpha, -\sum_i q_i \mathbf{r}_i \phi_\beta\right)\cdot\hat{\varepsilon}_\lambda \begin{Bmatrix} 1 \\ \\ \dfrac{\omega_\alpha - \omega_\beta}{\hbar\omega_\lambda} \end{Bmatrix} ,$$

$$G^\pm_\lambda(\alpha,\alpha) \equiv 0 ,$$

where $I_\lambda \equiv cN_\lambda\hbar\omega_\lambda/V$ is the intensity of the λth mode (cavity volume V). If the linear polarizations and a set of real ϕ_α are chosen, then $G^+_\lambda(G^-_\lambda)$ are real and symmetric (antisymmetric).

III. ADIABATIC EIGENVALUE PROBLEM

For calculation of transition probability, we use the level shifts and the coupling between states, obtained below by solution of the adiabatic eigenvalue problem. We write

$$b_{\{\nu_\lambda\}}(\alpha) \equiv e^{-i\int^y E(y')dy'} a_{\{\nu_\lambda\}}(\alpha) , \tag{3.1}$$

where $\left(E(y),\mathbf{a}\right)$ is the "adiabatic eigensolution" to be found. In the adiabatic limit

$$\frac{d}{dy} a_{\{\nu_\lambda\}}(\alpha) = 0 .$$

Use of these expressions in Eqs. (2.3) gives

$$E\, a_{\{\nu_\lambda\}}(\alpha) = W_{\{\nu_\lambda\}}(\alpha)\, a_{\{\nu_\lambda\}}(\alpha) + \sum_{\gamma,\beta} G^\pm_\gamma(\alpha,\beta) \times \left(a_{\{\nu_\gamma-1\}}(\alpha) \pm a_{\{\nu_\gamma+1\}}(\beta)\right) . \tag{3.2}$$

With $\left(E_{\{\nu_\lambda\}\alpha}, \mathbf{a}^{\{\nu_\lambda\}\alpha}\right)$ labeled such that as the interaction $G^\pm_\lambda(\alpha,\beta) \to 0$

$$E_{\{\nu_\lambda\}\alpha} \to W_{\{\nu_\lambda\}}(\alpha) ,$$

it can be shown from Eqs. (3.2) that if a particular $\left(E_{\{\rho_\lambda\}\alpha}, \underset{\sim}{a}^{\{\rho_\lambda\}\alpha}\right)$ is found, then $\left(E_{\{\rho_\lambda+\mu_\lambda\}\alpha}, a^{\{\rho_\lambda+\mu_\lambda\}\alpha}\right)$ for the same α but any $\{\mu_\lambda\}$ can be obtained by

$$E_{\{\rho_\lambda+\mu_\lambda\}\alpha} = E_{\{\rho_\lambda\}\alpha} - \mu_\lambda F_\lambda \quad ,$$

and

$$a^{\{\rho_\lambda+\mu_\lambda\}\alpha}_{\{\nu_\lambda\}}(\beta) = a^{\{\rho_\lambda\}\alpha}_{\{\nu_\lambda-\mu_\lambda\}}(\beta) \quad .$$

We proceed to solve Eqs. (3.2).

A. Single Field Mode

Converting Eqs. (3.2) into matrix notations and dropping the mode index, we let $\underset{\sim}{G}^{\pm}$ be the $n \times n$ matrix $\left[G^{\pm}(\alpha,\beta)\right]$, $\underset{\sim}{D}$ be the $n \times n$ diagonal matrix $\left[\left(E - W_\nu(\alpha)\right)\delta_{\alpha\beta}\right]$ and $\underset{\sim}{a}_\nu$ be the n-component column vector. Then Eqs. (3.2) become

$$\underset{\sim}{D}_\nu \underset{\sim}{a}_\nu = \underset{\sim}{G}^{\pm}(\underset{\sim}{a}_{\nu-1} \pm \underset{\sim}{a}_{\nu+1}) \, , \tag{3.3}$$

for all ν.

At y where an adiabatic level of interest (ρ,σ) is not in near resonance with any other levels, we let

$$a_\nu(\alpha) = d_\nu(\alpha)\, a_\rho(\sigma) \tag{3.4}$$

for all (ν,α). Thus $d_\rho(\sigma) = 1$. Upon substitution of Eqs. (3.4), and factorizing out $a_\rho(\sigma)$, Eqs. (3.3) become

$$\underset{\sim}{D}_\nu \underset{\sim}{d}_\nu = \underset{\sim}{G}^{\pm}(\underset{\sim}{d}_{\nu-1} \pm \underset{\sim}{d}_{\nu+1}) \quad . \tag{3.5}$$

Defining $\underset{\sim}{T}_\nu$ for $\nu > \rho$ by

$$\underset{\sim}{d}_\nu = \underset{\sim}{T}_\nu \underset{\sim}{d}_{\nu-1} \quad ,$$

We obtain directly from Eqs. (3.4) the recurrence relation for $\underset{\sim}{T}_\nu$,

$$\underset{\sim}{T}_\nu = (\underset{\sim}{D}_\nu \mp \underset{\sim}{G}^{\pm} \underset{\sim}{T}_{\nu+1})^{-1} \underset{\sim}{G}^{\pm} \quad . \tag{3.6a}$$

Since $D_\nu(\alpha,\beta)^{-1} \to (\nu F)^{-1} \delta_{\alpha\beta} \to 0$ as $\nu \to \infty$, $\underset{\sim}{T}_\nu$ has the limit

$$\underset{\sim}{T}_\nu \to (\nu F)^{-1} \underset{\sim}{G}^{\pm} \to 0 \quad . \tag{3.6b}$$

Similarly for $\nu < \rho$, we define $\underset{\sim}{T}'_\nu$ by

$$\underset{\sim}{d}_\nu \equiv \underset{\sim}{T}'_\nu \underset{\sim}{d}_{\nu+1}$$

and obtain from Eqs. (3.5)

$$\underset{\sim}{T}'_\nu = \pm(\underset{\sim}{D}_\nu - \underset{\sim}{G}^{\pm} \underset{\sim}{T}'_{\nu-1})^{-1} \underset{\sim}{G}^{\pm} \quad . \tag{3.7a}$$

As before

$$\underset{\sim}{T}'_\nu \to 0 \text{ as } \nu \to -\infty \quad . \tag{3.7b}$$

Alternatively, if $(\underset{\sim}{G}^{\pm})^{-1}$ exists, then defining $\underset{\sim}{U}^{\pm}_\nu$, $\underset{\sim}{H}_\nu$ and $\underset{\sim}{H}'_\nu$ by

$$\underset{\sim}{U}^{\pm}_\nu \equiv \underset{\sim}{D}_\nu^{-1} \underset{\sim}{G}^{\pm} \, ,$$

$$\underset{\sim}{T}_\nu \equiv \underset{\sim}{U}^{\pm}_\nu \underset{\sim}{H}_\nu \, , \qquad \nu > \rho$$

and

$$\underset{\sim}{T}'_\nu \equiv \underset{\sim}{U}^{\pm}_\nu \underset{\sim}{H}'_\nu \, , \qquad \nu < \rho$$

we obtain either directly from Eqs. (3.5) or from Eqs. (3.6) and (3.7), the relations

$$\underset{\sim}{H}_\nu = \left[\underset{\sim}{I} \mp \underset{\sim}{U}^{\pm}_{\nu+1} \underset{\sim}{H}_{\nu+1} \underset{\sim}{U}^{\pm}_\nu\right]^{-1} \, ,$$

with

$$\underset{\sim}{H}_{\nu} \to \underset{\sim}{I} \quad \text{as} \quad \nu \to \infty \; ; \tag{3.8}$$

and

$$\underset{\sim}{H}'_{\nu} = \pm\left[\underset{\sim}{I} - \underset{\sim}{U}^{\pm}_{\nu-1}\, \underset{\sim}{H}'_{\nu-1}\, \underset{\sim}{U}^{\pm}_{\nu}\right]^{-1} , \tag{3.9}$$

with $\underset{\sim}{H}'_{\nu} \to \pm\underset{\sim}{I}$ as $\nu \to -\infty$.

Thus with a cut-off value M, we let $\underset{\sim}{T}_{M+1} = 0$ and $\underset{\sim}{T}'_{-M-1} = 0$. Or if we take the approach using H-matrices, we let $\underset{\sim}{H}_{M+1} = \underset{\sim}{I}$, $\underset{\sim}{H}'_{-M-1} = \pm\underset{\sim}{I}$.[17] For a given physical system, the smallest value of M is determined according to the accuracy desired by running a few numerical tests. Starting from these limits we can generate all other $\underset{\sim}{T}_{\nu}\left(-M < \nu(\neq\rho) < M\right)$ by the recurrence relations. Thus $\underset{\sim}{d}_{\rho\pm1}$ can be expressed in terms of $\underset{\sim}{d}_{\rho}$. With $d_{\rho}(\sigma) = 1$ known, all other $(n-1)\, d_{\rho}(\alpha \neq \sigma)$ are obtained by solving the $n-1$ inhomogeneous linear equations obtained from the $\underline{(\nu = \rho)}$th set of Eqs. (3.5) with $\alpha \neq \sigma$.

Finally use of the $(\nu = \rho,\ \alpha = \sigma)$ equation of Eqs. (3.5) gives

$$E = W_{\rho}(\sigma) + \sum_{\beta,\gamma} G^{\pm}(\sigma,\beta)\left(T'_{\rho-1}(\beta,\gamma) \pm T_{\rho+1}(\beta,\gamma)\right) d_{\rho}(\gamma)$$

where $d_{\rho}(\sigma) = 1$ has been used. This equation is used to find the adiabatic eigenvalue $E_{\rho\sigma}$ by successive iteration, starting with trial value $W_{\rho}(\sigma)$. The second term on the right is the shift of the unperturbed level $W_{\rho}(\sigma)$. For low intensity, it agrees with the value given by perturbation theory.

Near y where two levels (ρ,σ) and (μ,τ) are nearly degenerate (for example, Fig. 1) we let

$$a_{\nu}(\alpha) = d_{\nu}(\alpha)\, a_{\rho}(\sigma) + s_{\nu}(\alpha)\, a_{\mu}(\tau) , \tag{3.10}$$

for all ν,α. It follows that

$$d_{\rho}(\sigma) = 1 = s_{\mu}(\tau) , \quad d_{\mu}(\tau) = 0 = s_{\rho}(\sigma) . \tag{3.11}$$

To find $\underset{\sim}{d}_{\nu}(\alpha)$'s, which are independent of $a_{\mu}(\tau)$, we substitute $d_{\nu}(\alpha)\, a_{\rho}(\sigma)$ in place of $a_{\nu}(\alpha)$ in Eqs. (3.3) and obtain equations the same as Eqs. (3.5). Thus all the $\underset{\sim}{d}_{\nu}$ can be found by the same procedure as before except for $\underset{\sim}{T}_{\mu}$ (assuming $\tau > \sigma$ and $\mu > \rho$). For $\nu = \mu$, since we cannot use the "singular" equation $(\nu = \mu,\ \alpha = \tau)$ to find the d's, $\underset{\sim}{T}_{\mu}$ is obtained (i) by filling its $\underline{(\alpha = \tau)}$th row by zeroes to satisfy $\underset{\sim}{d}_{\mu}(\tau) = 0$; and (ii) by directly inverting the rest of the $(n-1)$ equations with $\nu = \mu$ to obtain the other $(n-1)$ rows. The $s_{\nu}(\alpha)$'s are found similarly by substituting $s_{\mu}(\alpha)\, a_{\mu}(\tau)$ in place of $a_{\nu}(\alpha)$ in Eqs. (3.3). Defining $\underset{\sim}{V}_{\nu}$ by $\underset{\sim}{s}_{\nu} = \underset{\sim}{V}_{\nu}\underset{\sim}{s}_{\nu-1}$ for $\nu > \mu$ and $\underset{\sim}{V}'_{\nu}$ by $\underset{\sim}{s}_{\nu} = \underset{\sim}{V}'_{\nu}\underset{\sim}{s}_{\nu+1}$, we note that $\underset{\sim}{V}_{\nu} = \underset{\sim}{T}_{\nu}$ for $\mu < \nu \leq M$ and $\underset{\sim}{V}'_{\nu} = \underset{\sim}{T}'_{\nu}$ for $-M \leq \nu < \rho$.

Finally, the characteristic equation resulting from the (μ,τ) and (ρ,σ) equations of Eqs. (3.3) with substitution of Eqs. (3.10) for $a_{\rho\pm1}(\alpha)$ and $a_{\mu\pm1}(\alpha)$ has roots

$$E_\mu = \frac{1}{2}\left\{W'_\mu(\tau) + W'_\rho(\sigma) + \left[\left(W'_\mu(\tau) - W'_\rho(\sigma)\right)^2 + 4G^2\right]^{\frac{1}{2}}\right\} ,$$

$$E_\ell = \frac{1}{2}\left\{W'_\mu(\tau) + W'_\rho(\sigma) - \left[\left(W'_\mu(\tau) - W'_\rho(\sigma)\right)^2 + 4G^2\right]^{\frac{1}{2}}\right\} , \qquad (3.12)$$

where

$$W'_\rho(\sigma) \equiv W_\rho(\sigma) + \sum_\beta G^\pm(\sigma,\beta)\left(d_{\rho-1}(\beta) \pm d_{\rho+1}(\beta)\right) ,$$

$$W'_\mu(\tau) \equiv W_\mu(\tau) + \sum_\beta G^\pm(\tau,\beta)\left(s_{\mu-1}(\beta) \pm s_{\mu+1}(\beta)\right) ,$$

are the "shifted levels", and

$$G \equiv - \sum_\beta G^\pm(\sigma,\beta)\left(s_{\rho-1}(\beta) \pm s_{\rho+1}(\beta)\right)$$

$$= - \sum_\beta G^\pm(\tau,\beta)\left(d_{\mu-1}(\beta) \pm d_{\mu+1}(\beta)\right) ,$$

is the coupling constant. The last equality is based on hermiticity. We may use any one of the expressions of E_u and E_ℓ for iteration to find $(E_u, \underset{\sim}{a}^u)$ and/or $(E_\ell, \underset{\sim}{a}^\ell)$. The minimum of the level separation $E_u - E_\ell$ is the "point of closest approach" around which we calculate the transition probability.

B. Many Field Modes

For the sake of clarity, we will indicate the generalization of the above method to many field modes with the case of two modes. We shall use this in the numerical example in Sec. VB. Equations (3.2) for two field modes may be written in the form

$$\underset{\sim}{D}_{\nu_2} \underset{\sim}{a}_{\nu_2} = \underset{\sim}{G}^\pm(\underset{\sim}{a}_{\nu_2-1} \pm \underset{\sim}{a}_{\nu_2+1}) , \qquad (3.13)$$

where

$$\underset{\sim}{D}_{\nu_2}(\nu_1\alpha,\nu'_1\alpha') = (E + \nu_2 F_2)\delta_{\nu_1\nu'_1}\delta_{\alpha\alpha'} - \Big[W_{\nu_1}(\alpha)\,\delta_{\nu_1\nu'_1}\delta_{\alpha\alpha'}$$

$$+ G_1^\pm(\alpha,\alpha')(\delta_{\nu_1\nu'_1+1} \pm \delta_{\nu_1\nu'_1-1})\Big] ,$$

$$G^\pm(\nu_1\alpha,\nu'_1\alpha') \equiv \delta_{\nu_1\nu'_1}\, G_2^\pm(\alpha,\alpha')$$

and

$$a_{\nu_2}(\nu'_1\alpha') \equiv a_{\nu'_1\nu_2}(\alpha') .$$

For transition where photon number of only one mode changes, that mode should be assigned the role of mode 2 here. Equations (3.13) has the same form as Equations (3.3) for one mode. Thus the solution is similar as before, even though $\underset{\sim}{D}_{\nu_2}$ are not diagonal. For near resonance between levels $(\rho_1\rho_2\sigma)$ and $(\mu_1\mu_2\tau)$, for example, we write

$$a_{\nu_2}(\nu_1\alpha) \equiv d_{\nu_2}(\nu_1\alpha)\, a_{\rho_2}(\rho_1\sigma) + s_{\nu_2}(\nu_1\alpha)\, a_{\mu_2}(\mu_1\tau) , \qquad (3.14)$$

for all α, $-M_1 \leq \nu_1 \leq M_1$, and $-M_2 \leq \nu_2 \leq M_2$, and use component equations $(\rho_1\rho_2\sigma)$ and $(\mu_1\mu_2\tau)$ to obtain the corresponding characteristic roots as in Equations (3.12). The level shifts and the coupling constant contain additive contribution from each mode.

IV. A TRANSITION PROBABILITY FORMULA

Suppose we have (near) resonance occurring between two levels "1" and "2" near $y = 0$. Then only the probability amplitude b_1 and b_2 will vary significantly while all other $b_\nu(\alpha)$'s in Eqs. (2.3) may be approximated by their adiabatic counterparts given by

Eqs. (3.1). We may use either a^{μ} or a^{ℓ}, the resulting difference of such choice being small near the point of closest approach.[18] Substitution of Eqs. (3.1), Eqs. (3.10), or (3.14) into the component equations "1" and "2" of Eqs. (2.3) gives

$$i\frac{db_1}{dy} - W_1' b_1 = G b_2 ,$$

$$i\frac{db_2}{dy} - W_2' b_2 = G b_1 , \qquad (4.1)$$

where W_i' and G are respectively the shifted levels and coupling constant defined before. The boundary conditions are that in the remote past $b_2 = 0$ and $|b_1| = 1$. Now we derive a new formula useful when the shifted levels have two well-defined relative slopes b and b',

$$W_2' - W_1' = \begin{cases} a - by & y \leqslant 0 \\ a + b'y & y > 0 \end{cases}$$

where a is the minimum level separation. See Fig. 1. The only drawback in the above approximation is the introduction of discontinuity of slope of $W_2' - W_1'$ at $y = 0$. However, the advantage is that without further approximation a transition probability formula can be derived rigorously and is applicable even when $a = 0$.

We introduce, with $k_1 \equiv \int_0^y W_1'(y')dy'$,

$$b_2(y) = U(y)\, e^{-ik_1} e^{-i\frac{a}{2}y} \begin{Bmatrix} e^{i\frac{b}{4}y^2} \\ e^{-i\frac{b'}{4}y^2} \end{Bmatrix} , \text{ for } \begin{matrix} y \leqslant 0 \\ y > 0 \end{matrix}$$

into Eq. (4.1) and obtain, neglecting the small dependency of G on y,[19]

$$\frac{d^2U}{dy^2} + \left[G^2 - \frac{ib}{2} + \frac{b^2}{4}\left(y - \frac{a}{b}\right)^2\right]U = 0 , \qquad y \leqslant 0$$

$$\frac{d^2U}{dy^2} + \left[G^2 + \frac{ib'}{2} + \frac{b'^2}{4}\left(y + \frac{a}{b'}\right)^2\right]U = 0 , \qquad y > 0 .$$

We need to find only the solution $U(y;\, |a|, |b|, |b'|)$; for the solution $U(y;\, -|a|, -|b|, -|b'|) = U^*(y, |a|, |b|, |b'|)$ as can be shown easily from the above equations.

Now with definitions

$$z_- \equiv \left(y - \frac{a}{b}\right) b^{\frac{1}{2}} e^{-i\frac{\pi}{4}} \equiv y_- b^{\frac{1}{2}} e^{-i\frac{\pi}{4}}$$

$$z_+ \equiv \left(y + \frac{a}{b'}\right) b'^{\frac{1}{2}} e^{-i\frac{\pi}{4}} \equiv y_+ b'^{\frac{1}{2}} e^{-i\frac{\pi}{4}} ,$$

the above equations are reduced to the Weber's equations[20]

$$\frac{d^2U(z_-)}{dz_-^2} + \left[n + \frac{1}{2} - \frac{z_-^2}{4}\right]U(z_-) = 0 \qquad y \leqslant 0$$

$$\frac{d^2U(z_+)}{dz_+} + \left[n' + \frac{1}{2} - \frac{z_+^2}{4}\right]U(z_+) = 0 \qquad y > 0$$

where $n \equiv ip$, $\qquad n' = ip' - 1$,

$$p \equiv \frac{G^2}{b} , \qquad p' \equiv \frac{G^2}{b'} .$$

For $y < 0$, the solution satisfying the boundary condition $b_2 = 0$ and $|b_1| = 1$ in the "remote past" $\left(\text{i.e., } |b^{\frac{1}{2}}y - \frac{a}{b^{\frac{1}{2}}}| \gg 1\right)$ is

$$U(z_-) = \frac{|G|}{b^{\frac{1}{2}}} e^{-\frac{\pi}{4}p} D_{-n-1}(-iz_-) \quad .$$

A general solution for $y > 0$ is

$$U(z_+) = L\, D_{-n'-1}(-iz_+) + M\, D_{n'}(-z_+)$$

where L and M are coefficients to be determined by demanding continuity of b_2 and of its slope at $y = 0$. We obtain

$$L = N\left[n' e^{i\frac{3\pi}{4}} D_1 D_6 - (b/b')^{\frac{1}{2}}(n+1) e^{i\frac{\pi}{4}} D_2 D_5\right]/D \ ,$$

$$M = N\left[(a/b'^{\frac{1}{2}}) e^{i\frac{\pi}{2}} D_1 D_3 - (n'+1) e^{i\frac{\pi}{4}} D_1 D_4 + (b/b')^{\frac{1}{2}}(n+1) \times e^{i\frac{\pi}{4}} D_2 D_3\right]/D \ ,$$

where

$$N \equiv |G| b^{-\frac{1}{2}} e^{-\frac{\pi}{4}p} \ ,$$

$$D \equiv (a/b'^{\frac{1}{2}}) e^{i\frac{\pi}{2}} D_3 D_5 + n' e^{i\frac{3\pi}{4}} D_3 D_6 - (n'+1) e^{i\frac{\pi}{4}} D_4 D_5 \ ,$$

$$D_1 \equiv D_{-n-1}\left(\frac{a}{b^{\frac{1}{2}}} e^{i\frac{\pi}{4}}\right) \ ,$$

$$D_2 \equiv D_{-n-2}\left(\frac{a}{b^{\frac{1}{2}}} e^{i\frac{\pi}{4}}\right) \ ,$$

$$D_3 \equiv D_{-n'-1}\left(\frac{a}{b'^{\frac{1}{2}}} e^{-i\frac{3\pi}{4}}\right) \ ,$$

$$D_4 \equiv D_{-n'-2}\left(\frac{a}{b'^{\frac{1}{2}}} e^{-i\frac{3\pi}{4}}\right) \ ,$$

$$D_5 \equiv D_{n'}\left(\frac{a}{b'^{\frac{1}{2}}} e^{i\frac{3\pi}{4}}\right) \ ,$$

$$D_6 \equiv D_{n'-1}\left(\frac{a}{b'^{\frac{1}{2}}} e^{i\frac{3\pi}{4}}\right) \ .$$

For $|z_+| = b'^{\frac{1}{2}} y + \frac{a}{b'^{\frac{1}{2}}} \gg 1$, we obtain the asymptotic formula for $b_2(y)$

$$b_2(y) = \left(L\, e^{-\frac{3\pi}{4}p'} + M \frac{(2\pi)^{\frac{1}{2}}}{\Gamma(1 - ip')} e^{-\frac{\pi}{4}p'}\right) \times \exp\left[-i\int_0^y W_2'\, dy' - i\frac{p'}{2} \ln b'\left(y + \frac{a}{b'}\right)^2 - \frac{ia^2}{4b'}\right] \ ,$$

from which the transition probability is

$$|b_2|^2 = \left|L\, e^{-\frac{3\pi}{4}p'} + M \frac{(2\pi)^{\frac{1}{2}}}{\Gamma(1 - ip')} e^{-\frac{\pi}{4}p'}\right|^2 \ . \qquad (4.2)$$

For the special case of $a/b^{\frac{1}{2}} \gg 1$ and $a/b'^{\frac{1}{2}} \gg 1$, we obtain

$$|b_2|^2 = \left[\frac{\sqrt{2}\,|G|}{a}\left(1 - e^{-2\pi p'}\right)\right]^2 ,$$

which has desirable behavior with respect to a, G and b', but is independent of b.

V. NUMERICAL EXAMPLES

The orbit of the relative motion of the colliding atoms enter into our calculation through (i) $G(\alpha,\beta)$ which depends on the orientation of the internuclear axis relative to the polarization vector $\hat{\epsilon}$; and (ii) the velocity dR/dt in the relative slopes

$$\alpha_{ij} = \frac{a_o}{v}\frac{d}{dR}(W_i' - W_j')\frac{dR}{dt}$$

that occur in the transition probability formulae. To take (i) into consideration, we integrate the differential orbit equation, using the unperturbed potential curves as a good approximation (though strictly speaking, the self-consistent shifted potential curves should be used). The evaluation of dR/dt, final transition probability and cross section have been discussed in KW.

A. Li and H Scattering

As examples, we consider scattering of Li with H in an intense field mode with $\lambda = 0.826\ \mu$ in the geometry of Fig. 2 for relative speed $v_\infty = 5 \times 10^5$ cm/sec. Values of $w_\alpha(R)$ and $(\phi_\alpha, \sum_i r_{mi}\cdot\phi_\beta)$ for the lowest lying singlet states $X^1\Sigma^+$, $A^1\Sigma^+$, and $B^1\Pi$ are taken from Docken and Hinze.[21] These three levels represent a fairly good approximation because according to the less accurate calculation of Bender and Davidson,[22] the higher levels all lie at least about one-photon ($\hbar\omega \approx 1.5$ eV) energy above the $B^1\Pi$ level. We assume that the atoms are initially in the electronic singlet ground state $X^1\Sigma^+$. There are 1-, 2-photon resonant transition to $A^1\Sigma^+$ near internuclear separations $9.4\ a_0$ and $5.9\ a_o$, and $3.7\ a_o$ respectively; while 2-, 3-photon resonant transitions to $B^1\Pi$ occur at $5.1\ a_o$ and $3.1\ a_o$ respectively. Using

a Landau-Zener-like formula,[10] we calculated the transition probabilities to these two excited states and the cross sections are presented in Fig. 3. We observe that only for low enough intensity are the cross sections proportional to I and I^2 for transitions to $A^1\Sigma^+$ and $B^1\Pi$ respectively, as expected from perturbation theory.

B. Stimulated Emission by Field-dependent Lowering of Potential Barrier[23]

We consider here colliding atoms with model adiabatic potential curves and dipole matrix elements illustrated in Fig. 4. Initially the quasi-molecule is in state 2, which has a potential barrier (e.g., due to avoided crossing) at R_b. For $R > R_b$, the dipole transition to the state 1 is forbidden, while for $R < R_b$ it is allowed. For diatomics, the initial state 2 is achieved by some pumping. But for polyatomics, no pumping is necessary because the potential surface 2 may represent ground state of one configuration of the quasi-molecular complex while potential surface 1 corresponds to a rearranged configuration. At thermal relative velocities, the potential barrier is too high for the classical penetration (or too little quantum mechanical tunnelling) into region $R < R_b$. One way to overcome the barrier is to lower it by a sufficiently intense laser field with a photon energy smaller than the energy gap between level 2 and 3 in the neighborhood of R_b. Once the quasi-molecule penetrates into the $R < R_b$ region, it will most likely radiate near the classical turning point R_c at a second frequency. For case depicted in Fig. 4, part of the electronic energy upon photoemission is converted into relative kinetic energy of the colliding particles.

For numerical study, we consider the following unperturbed potential energy curves $w_i(R)$ (in eV) and dipole matrix elements (in AU) $M_{jk} = \left\langle j \middle| \sum_i \underset{\sim}{r}_i \middle| k \right\rangle$ for a model diatomic colliding system (of reduced mass = 20 × proton mass and relative speed = 10^5 cm/sec),

$$w_1(R) = 1.5\, e^{-5(R-3)} ,$$

$$w_2(R) = 2.9 + 0.1\left(1 - e^{-1.6(R-3.75)}\right)^2 + 0.153\, e^{-4.0(R-4.8)^2} ,$$

$$w_3(R) = 4.285 + 1.2\, e^{-3(R-3)} ,$$

$$M_{12}(R) = 3.033\, e^{-1.515(R-3.2)^2} ,$$

$$M_{23}(R) = 4\, e^{-0.738(R-4.85)^2} ,$$

$$M_{13}(R) = 2.0 ,$$

for $R \geq 3.2\ a_o$. We assume electronic state 2 and 3 to have the same Λ-quantum number while that of state 1 differs from theirs by 1. Scattering geometry is shown in Fig. 2, where $\hat{\varepsilon}$ is the linear polarization $\hat{\varepsilon}_1$ of the intense mode. The original barrier is 0.02 eV too high for classical penetration. With high-intensity laser field wavelength chosen to be 1.0648 μ, the bump is lowered by about 0.04 eV at $I_1 = 10^{12}$ W/cm^2. The colliding atoms for certain range of impact parameter ($b = 0$ to $b_{max} = 1.97\ a_o$) can now penetrate into the $R < R_b$ region. The system has certain probability $P_{I_2}(b)$ to radiate near R_c by stimulated emission into the second mode $\lambda_2 \approx 0.486\ \mu$ ($\hat{\varepsilon}_2$ is chosen parallel to $\hat{\varepsilon}_1$). The cross sections for stimulated emission into the second mode

$$\sigma_{I_2} = 2\pi \int_0^{b_{max}} db\ b\ P_{I_2}(b)$$

are given in Table I, $P_{I_2}(b)$ being approximated by formula (4.2). We assume that transition between the shifted levels 2 and 3 near R_b is negligible, because of the large off-resonance 0.06 eV. Raising the third level by 0.4 eV higher changes the amount of potential barrier down-shift by less than 10%. Thus the selection of the third level (or high intensity laser wavelength) is not severely restrictive.

An interesting effect occurs which is due to the fact that the coupling between the second and third levels depends on the angle between the internuclear axis of the colliding atoms and the space-fixed linear polarization $\hat{\varepsilon}_1$. Thus collisional systems with impact parameter $b = 1.46$ to $1.97\ a_o$ can get into the region $R < R_b$ but become bound due to the change of this angle on the outgoing trip. Values for cross section for such "trapping"

$$b_t = 2\pi \int_{1.46}^{1.97} db\ b\left(1 - P_{I_2}(b)\right),$$

are given in Table I. The trapped colliding system will become a "vibrating" molecule that keeps on rotating relative to $\hat{\varepsilon}_1$. Following approximately the motion of the bound molecule in the intense beam shows that after five vibrations, the atoms are separated again. But while bound, they radiate predominantly near R_c, thus enhancing σ_{I_2} by an amount $\Delta\sigma$ indicated in the last column of Table I.

VI. TRANSITION DUE TO INTENSITY VARIATION

Intense laser field in experiments is often pulsed. The above theory can be adapted to treat transition in an isolated atom being irradiated by an intense laser pulse.[24] Now the amount of level shift of the atom is a function of intensity of the pulse, which is in turn a function of time. For a particular atomic system with proper choice of the laser λ, one may get two pseudocrossings (PC) per pulse as shown in Fig. 5. The final transition probability per pulse (assumed symmetric) is given by

$$f = 2T(1 - T)$$

where $T = 1 - \exp(-2\pi G^2/|\alpha|)$ is the transition probability[10] at one PC. The relative slope between the two shifted levels W_2' and W_1'.

$$\alpha = -\frac{a_o}{v}\frac{d(W_2' - W_1')}{dI}\frac{dI}{dt} .$$

is evaluated at the "critical intensity" I' at which the point of closest approach of the adiabatic eigenlevels occurs.

The analysis below shows that desirable transition probability per pulse can be achieved by choice of pulse shape and pulse parameters. This may have important application in efficient optical pumping and in isotope separation. For example, to attain the maximum value $f = \frac{1}{2}$, the temporal slope of the pulse at I' is given by

$$\left|\frac{dI}{dt}\right| = 2\pi\delta/\ln 2$$

where

$$\delta \equiv G^2 \Big/ \left|\frac{a_o}{v}\frac{d(W_2' - W_1')}{dI}\right| . \qquad (6.1)$$

For a gaussian pulse, $I(t) = I_o e^{-t^2/\tau^2}$,

$$\left|\frac{dI}{dt}\right|_{I'} = 2I'(\ln I_o/I')^{\frac{1}{2}}/\tau .$$

This implies that for given δ and I', there is a pair of optimum values (I_o', τ') such that f equals $\frac{1}{2}$. They are related by

$$\tau' = 7.516 \times 10^{-23}(\ln 2)\, I'(\ln I_o'/I')^{\frac{1}{2}}/\pi\delta$$

where δ is in atomic units, I' in watt/cm^2 and τ' in second. The validity condition for applying the transition probability formula for T above requires that for a gaussian pulse,

$$2.4 \times 10^5\, \delta\, I'\tau\left(\ln(I_o/I')\right)^{3/2} \gg 1$$

is to be satisfied.

As examples, we have calculated the quantities δ, I' characterizing the 2-photon transition from ground state 3s to 5s of sodium atom and the 8-photon transition from ground 2s state to 3s state of lithium atom. (Table II.) From these values, transition probability for any pulse may be calculated. States of 3-8s, 3-5p, 3-5d, and 4-6f are included in the calculation for sodium atom; while states of 2-7s, 2-4p, 3-5d, and 4-6f of lithium atom are used. The energy levels are taken from Moore.[25] The magnitude of dipole matrix elements are calculated from work of Anderson and Zilitiz[26] and their signs from Bates and Damgaard.[27] The range of wavelengths in the sodium case is chosen such that the 3s and 5s levels are shifted into 2-photon resonance because the 3p levels repel the 3s level stronger than they pull the 5s level. In the lithium case, the 3p

states strongly shift the 3s level down into 8-photon resonance with the 2s ground state.

It is found that for soidum atom irradiated by a gaussian pulse of $\lambda = 0.602396\ \mu$, $I_o = 6 \times 10^8\ W/cm^2$ and $\tau = 1$ nsec, final transition probability $f = \frac{1}{2}$ is closely attained. Figure 6 shows the sensitivity of f to τ over a range of wavelength. As can be shown from analytic expressions above, the result is not so sensitive to peak intensities.

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FOOTNOTES AND REFERENCES

* This work was supported by the U. S. Air Force Office of Scientific Research, under Grant No. 74-2716 and by the U. S. Energy Research and Development Administration.

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18. The difference in the final results in this paper due to the use of a_m^u or a_m^ℓ usually runs from 0.01% for intensity 10^8 W/cm^2 to less than 5% for 10^{12} W/cm^2. Alternatively, one may take the mean results.

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Table I. Cross sections of stimulated emission σ_{I_2}, of trapping σ_t, and of enhancement $\Delta\sigma$ as a function of the intensity of stimulated emission I_2.

$I_2(W/cm^2)$	$\sigma_{I_2}(a_o^2)$	$\sigma_t(a_o^2)$	$\Delta\sigma(a_o^2)$
1×10^7	1.14×10^{-3}	5.5	4.1×10^{-3}
4×10^7	4.54×10^{-3}	5.5	1.6×10^{-2}
1×10^8	1.14×10^{-2}	5.5	4.1×10^{-2}
4×10^8	4.54×10^{-2}	5.5	1.6×10^{-1}
1×10^9	1.13×10^{-1}	5.5	4.1×10^{-1}
4×10^9	4.51×10^{-1}	5.5	1.6
7×10^9	7.86×10^{-1}	5.5	2.9
1×10^{10}	1.12	5.5	4.1

Table II. Values of critical intensity (I') and pulse-independent factor (δ) in Eq. (6.1) for wavelengths (λ) considered in (a) 2-photon transition in sodium (3s → 5s); and (b) 8-photon transition in lithium (2s → 3s).

(a) Na			(b) Li		
$\lambda(\mu)$	$I'(W/cm^2)$	δ(a.u.)	$\lambda(\mu)$	$I'(W/cm^2)$	δ(a.u.)
6.02396E-01	1.00E+07	1.851E-17	2.94060E+00	1.90E+07	3.414E-49
6.02395E-01	1.74E+07	5.605E-17	2.94075E+00	5.30E+07	1.264E-45
6.02394E-01	2.55E+07	1.204E-16	2.94100E+00	1.10E+08	4.421E-43
6.02392E-01	4.00E+07	2.962E-16	2.94150E+00	2.30E+08	1.658E-40
6.02390E-01	5.50E+07	5.601E-16	2.94200E+00	3.50E+08	4.920E-39
6.02385E-01	9.50E+07	1.671E-15	2.94250E+00	4.70E+08	5.487E-38
6.02380E-01	1.35E+08	3.375E-15	2.94300E+00	6.00E+08	3.656E-37
6.02370E-01	2.10E+08	8.171E-15	2.94350E+00	7.30E+08	1.951E-36
6.02360E-01	2.90E+08	1.558E-14	2.94400E+00	8.65E+08	7.714E-36
6.02350E-01	3.75E+08	2.606E-14	2.94450E+00	1.00E+09	2.560E-35
6.02330E-01	5.25E+08	5.111E-14	2.94500E+00	1.14E+09	7.443E-35

FIGURE CAPTIONS

Fig. 1. Shifted level structure for which a transition probability formula is derived.

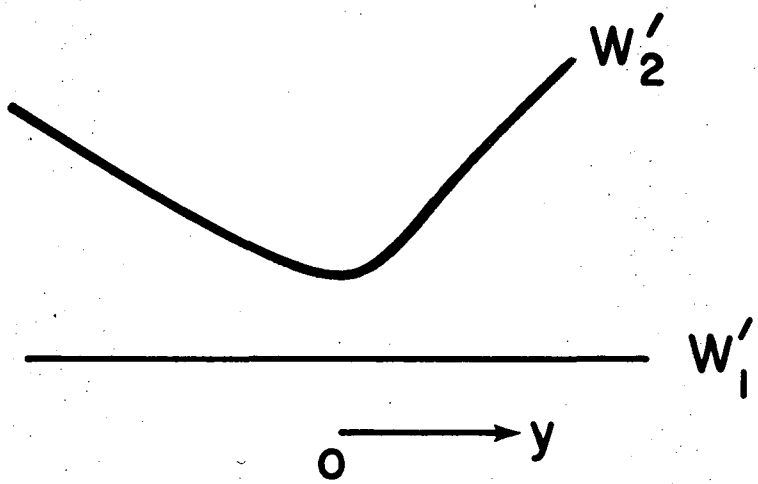
Fig. 2. Particular geometry for near-adiabatic scattering of two atoms in intense field. Internuclear axis is along $\hat{k}$ and the linear polarization $\hat{\varepsilon}$ is in the $\hat{i} - \hat{k}$ plane.

Fig. 3. Inelastic cross sections for the process $Li + H(X^1\Sigma^+) + \nu\hbar\omega \rightarrow Li + H(A^1\Sigma^+$ or $B^1\Pi)$ over a range of field intensity ($\lambda = 0.826\ \mu$).

Fig. 4. Potential curves and dipole matrix elements of model quasi-molecule for study of field-dependent lowering of potential barrier.

Fig. 5. Unperturbed atomic energy levels W_1 and W_2 are shifted into multiphoton resonances at critical intensity I' of the intense laser pulse.

Fig. 6. Transition probability per pulse, f, in $Na(3s) + 2\hbar\omega$ $Na(3s) + 2\hbar\omega \rightarrow Na(5s)$ at several wavelengths for a few gaussian pulses of the same peak intensity 6×10^8 W/cm^2.



XBL756-3338

Fig. 1

0 0 0 0 4 4 0 1 3 8 2

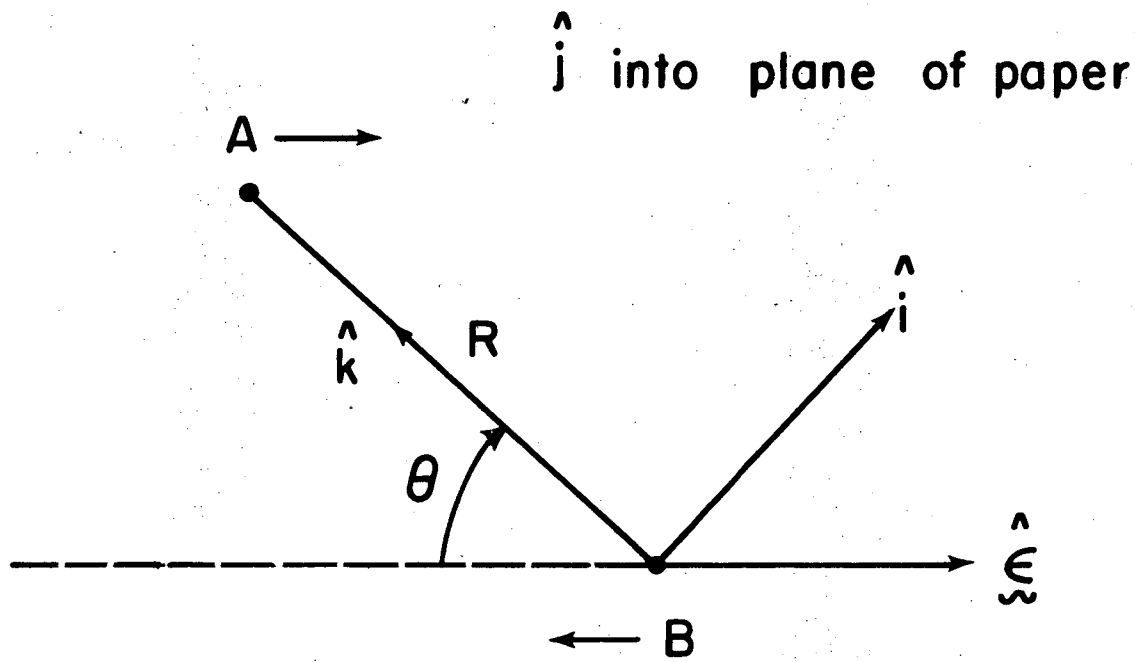


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Fig. 2

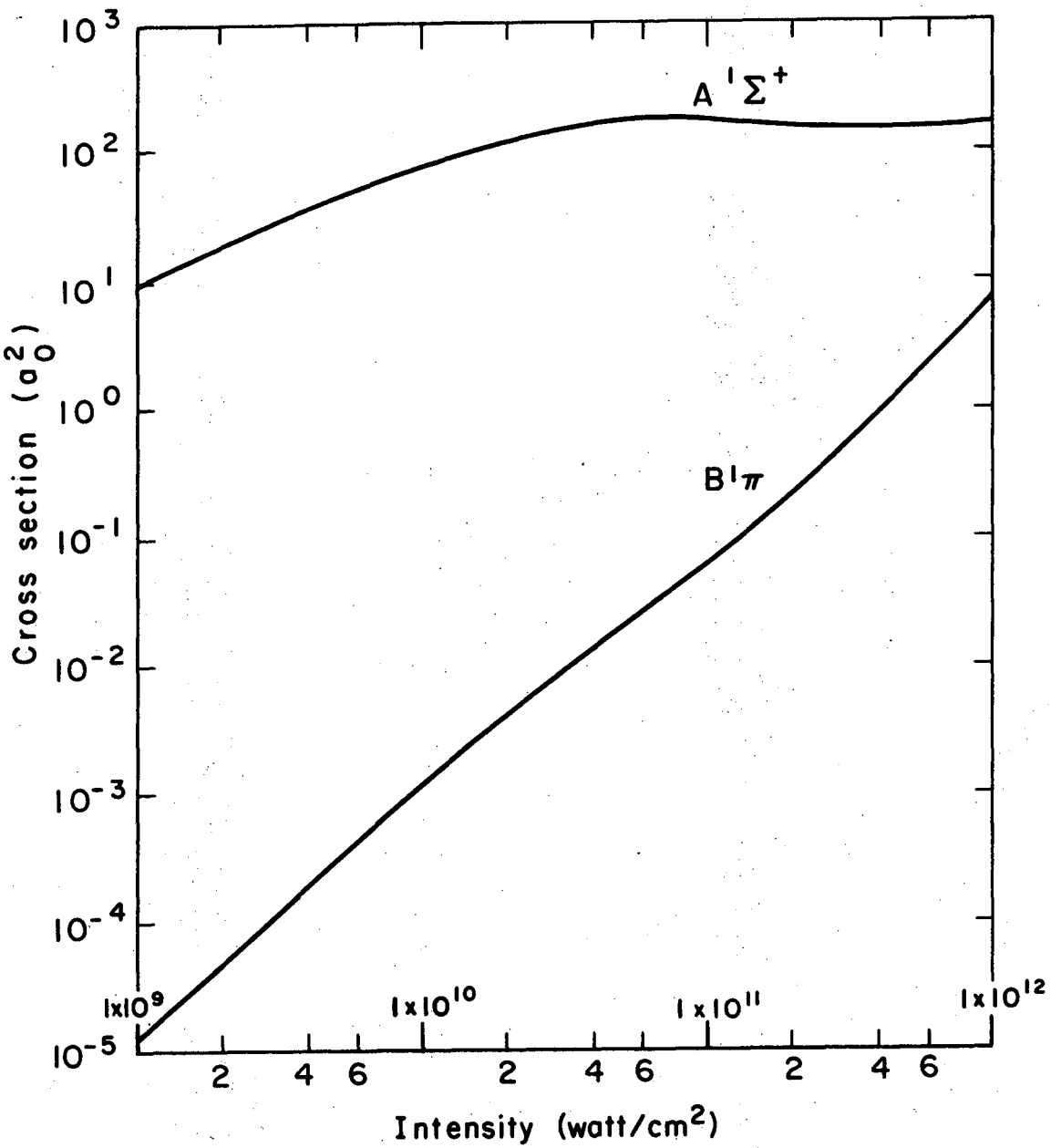

Fig. 3

0 0 0 0 4 4 0 1 3 8 3

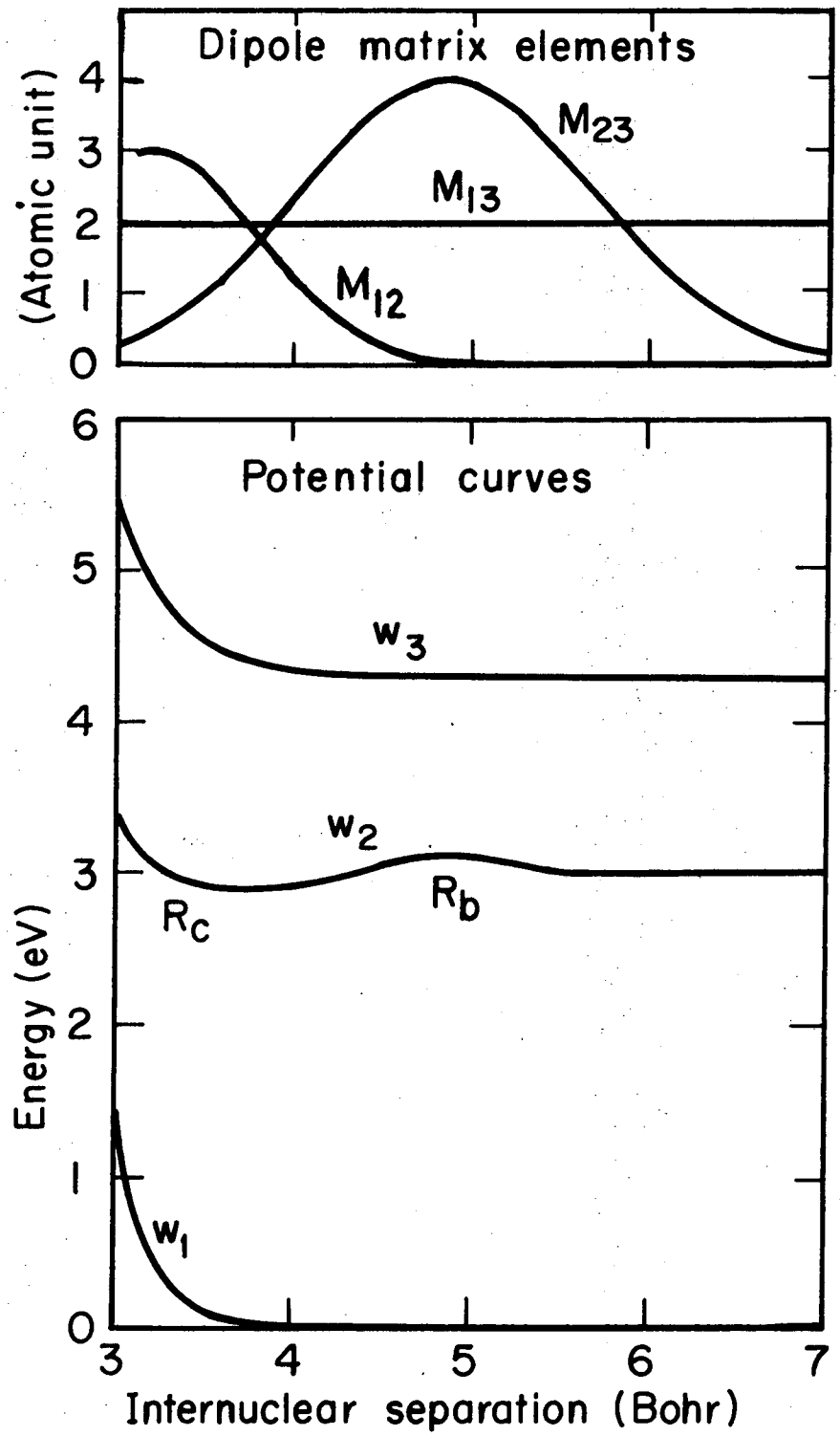

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Fig. 4

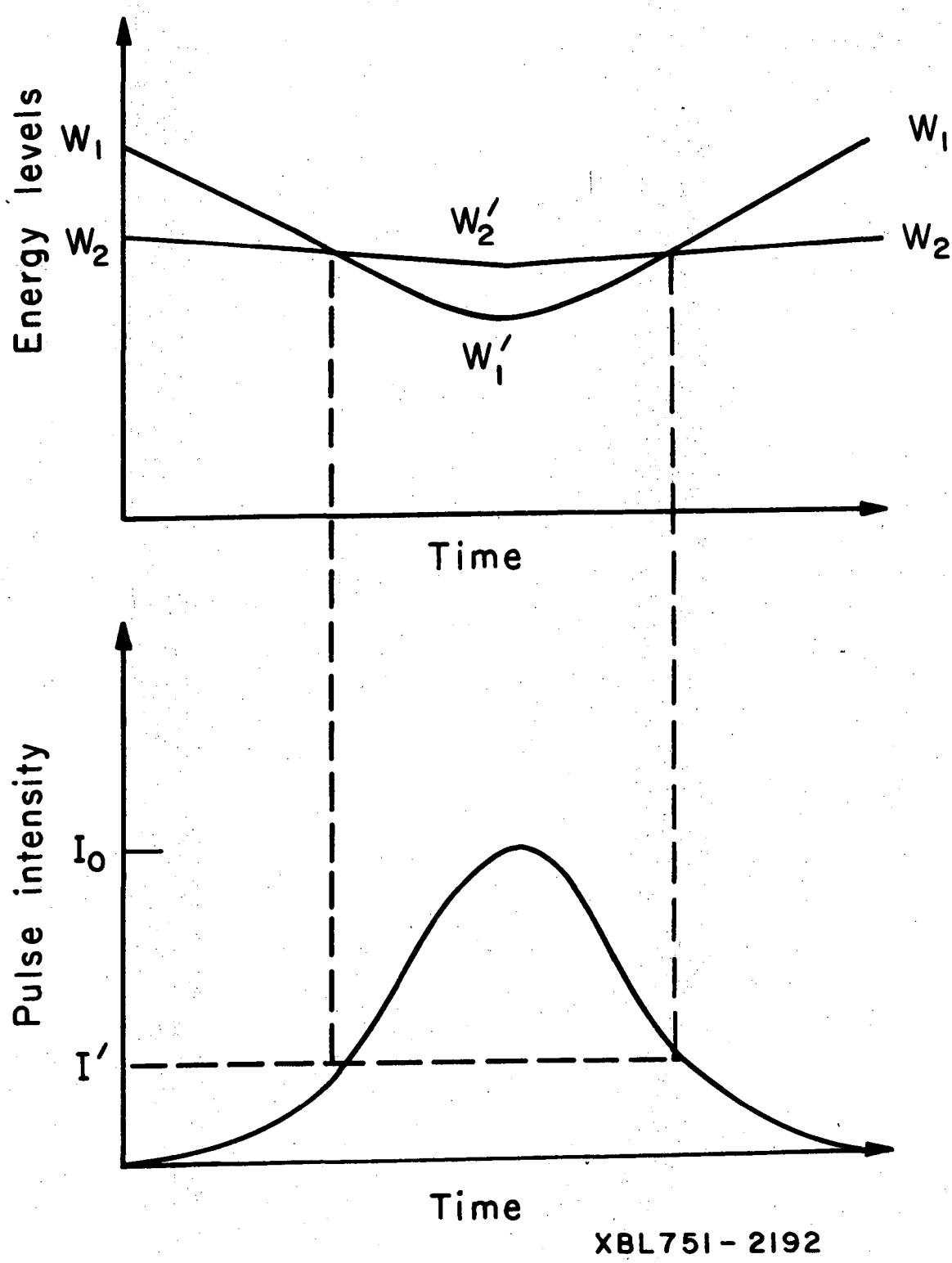
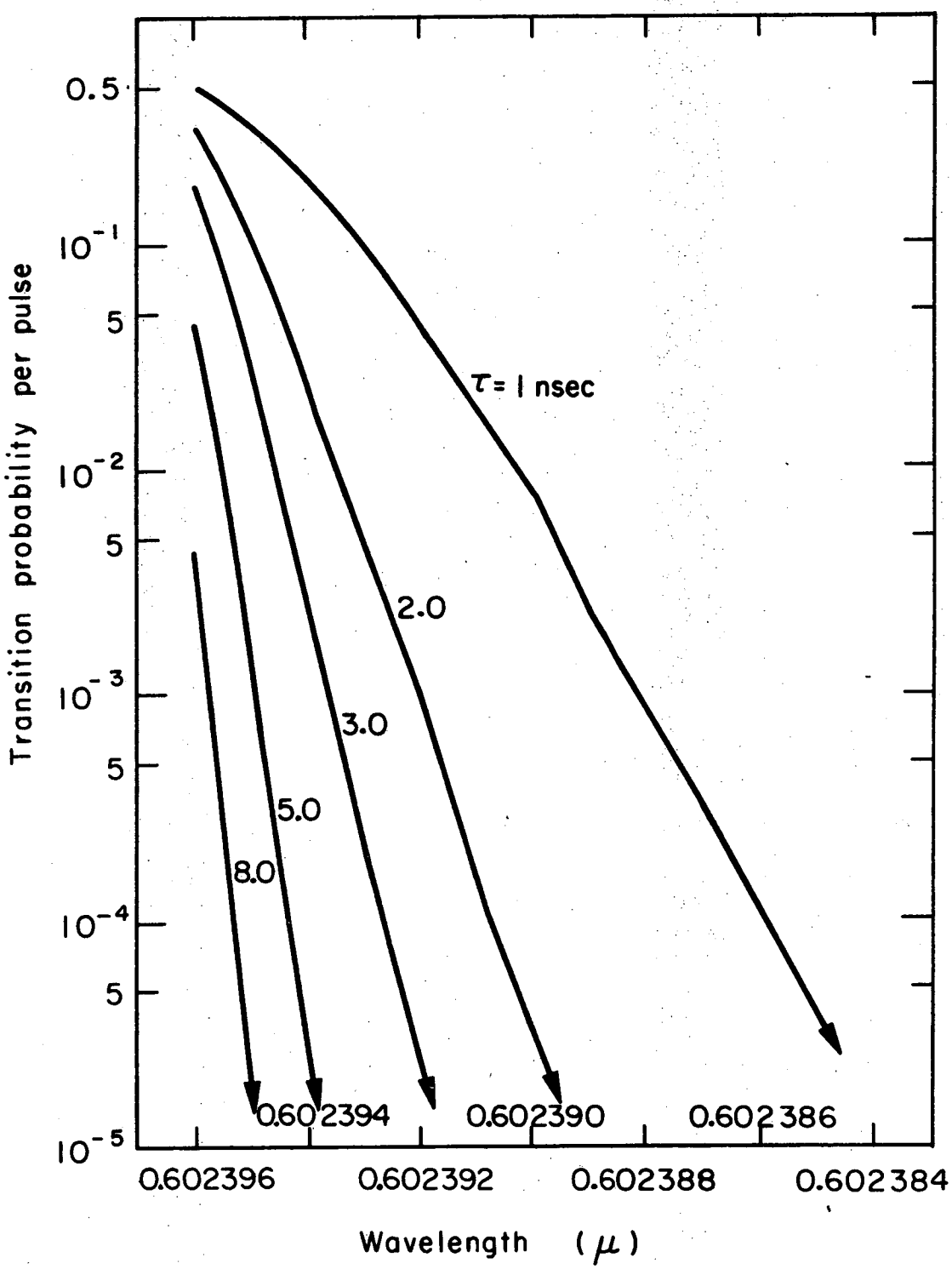


Fig. 5

0 0 0 0 4 4 0 1 3 8 4



XBL 751-2193

Fig. 6

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