Transition process under intense laser field: $\mathrm{Sr}(5s\,5p\ {}^1P_1)+\mathrm{Ca}(4s^2\ {}^1S_0)+\hbar\omega \rightarrow \mathrm{Sr}(5s^2\ {}^1S_0)+\mathrm{Ca}(3d\,4p\ {}^1F_3)$

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A theory for multipole-multipole collision under intense laser field is studied, and the formulas of transition probability and cross section are given. An application of this theory is made on the process given in the title.

I. INTRODUCTION

Since the pioneering work of Gudzenko *et al.* and the related experiments were published, a large amount of works have been presented on the laser-induced collision energy transfer (LICET).[1-20] The processes of LICET are classified as inelastic collision, charge transfer, a pair absorption, and radiative collisional fluorescence. They are followed by experiments and are summarized by Harris and collaboration.[14] Meanwhile, high-resolution experiments have been made by Brechignac *et al.*[16] on a sodium-strontium system and by Debarre[17] for a sodium-calcium mixture. The line profile of LICET experiments are proportional to the -0.8 power of the detuning frequency when collisional dipole-dipole interaction is decisive, although the theoretical result is -0.5. This discrepancy has not been solved yet. The theoretical studies of LICET are so far classified roughly into the two approaches, i.e., two-state and multiple- (mostly three-) state schemes. The former[11] is reduced to the celebrated Landau-Zener formula, perturbation theory,[8,9] and Gudzenko *et al.*'s method,[10] which give the formula of the cross section covering the whole range of the intensity of a laser field. Most of the works concerning the latter case are so made as to reduce the number of states to two.[4,10,13] Harris *et al.*[4] discussed by numerical analysis the dependence of the laser field and detuning frequency on the excitation cross section. However, the validity of the state truncation procedure remains indeterminate. Recent works of both experiment and theory on laser-assisted collision are collected by Picque[20] *et al.* George and collaborators[20] summarized semiclassical and quantum-mechanical approaches for chemical reactions and proposed the theory which combines radiative resonance formation with predissociation. Berman[20] made a study of the inelastic collision in a two-state scheme where the virtual states are considered in the forms of coupling operators. Polarization effects in energy transfer between laser-excited atoms are investigated by Nieuhuis[20] by means of density matrix. The density matrix was also used by Faisal[20] to analyze radiative Coulomb scattering and other processes in strong field.

In the previous report[18] (hereafter known as paper I) we suggested a method to solve three-channel equations and applied it to strontium-calcium collision where dipole-dipole interaction is predominant. Here we develop LICET processes in a high-intensity laser field and derive the formula of the cross section for prototype system. Also discussed is the effect of the detuning frequency.

II. THEORY

In considering the laser-induced transition, we adopt a prototype system,[3,4] i.e., a three-state model where each state is the product of the atomic wave functions of the colliding atoms. We wish to avoid a redundant repetition of the theory developed previously.[18] The brief review of the theory is as follows.

The system under consideration is simplified to the three states, $|f_1\rangle=|a_2\rangle|b_1\rangle$, $|f_2\rangle=|a_1\rangle|b_2\rangle$, and $|f_3\rangle=|a_1\rangle|b_3\rangle$, where $|a_j\rangle$ is the jth atomic state of isolated atom A and $|b_j\rangle$ is that of atom B as diagrammed in Fig. 1. Those atomic states are those of isolated atoms, i.e., both atoms are infinitely separated. The transition between $|f_1\rangle$ and $|f_2\rangle$ is via a multipole-multipole interaction but $|f_2\rangle$ and $|f_3\rangle$ by a laser field. The direct coupling between $|f_1\rangle$ and $|f_3\rangle$ is forbidden. The interaction Hamiltonian is

$$H_{\mathrm{int}}=(Y_A-Y_B)eE\cos(\omega t)+R^{-n}g^n(X'_A,X'_B)\ , \qquad (1)$$

where $E\cos(\omega t)$ is the laser field, R is the nuclear distance between A and B and the coordinate system and is the same as in paper I (Ref. 18). A linear trajectory is assumed for the nuclear motion throughout this paper. The wave function of the system is the linear combination of those three states

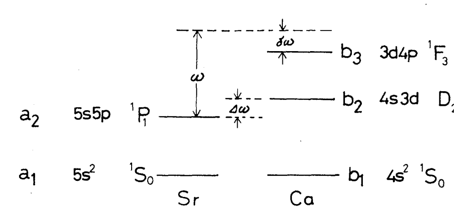

FIG. 1. Energy diagrams of Sr and Ca.

$$\Psi=\sum_{k=1}^{3} C_k\,|f_k\rangle e^{-i\omega_k t}\,, \tag{2}$$

where C_k is a coefficient and ω_k is the nth energy divided by $\hbar$. The coupled equations with respect to C_k under the boundary condition $C_n(-\infty)=\delta_{1n}$ are

$$\frac{d}{dt}\begin{bmatrix} C_1\\ C_2\\ C_3\end{bmatrix}=i\begin{bmatrix} 0 & u_{12} & 0\\ u_{21} & 0 & u_{23}\\ 0 & u_{32} & 0\end{bmatrix}\begin{bmatrix} C_1\\ C_2\\ C_3\end{bmatrix} \tag{3}$$

with

$$u_{12}=u_{21}^*=\hbar^{-1}R^{-n}g_{12}^n\exp(i\,\Delta\omega\,t)\,,$$

$$u_{23}=u_{32}^*=\frac{\mu^{B_2}E}{2\hbar}\exp(\Delta\omega+\delta\omega)t\,,$$

$$g_{12}^n=\langle f_1\,|\,g^n\,|\,f_2\rangle\,.$$

$\delta\omega$ is the detuning frequency and μ^{B_2} is the dipole moment between $|\,b_2\rangle$ and $|\,b_3\rangle$.

The S matrix only between $|f_2\rangle$ and $|f_3\rangle$ is

$$S=\begin{bmatrix} S_{22} & -S_{32}^*\\ S_{32} & S_{22}^*\end{bmatrix}$$

where

$$S_{22}=\frac{1}{2\alpha}e^{i\beta t/2}[(\alpha-\beta)e^{i\alpha t/2}+(\alpha+\beta)e^{-i\alpha t/2}],\quad S_{32}=\frac{u_{23}}{\alpha}e^{-i\beta t/2}(e^{i\alpha t/2}-e^{-i\alpha t/2}),$$
$$\alpha=[\beta^2+(2u_{23})^2]^{1/2},\ \text{and}\ \beta=\Delta\omega+\delta\omega\,. \tag{4}$$

Leaving aside the details of the previous report, we show the exact transition probabilities and related quantities

$$|\,C_1(t)\,|^2=\left|\exp\left[-i\int_{-\infty}^{t}\delta\,dt'\right]\right|^2$$
$$=1-D^\dagger D=1-|\,C_2\,|^2-|\,C_3\,|^2\,, \tag{5}$$

$$D(t)=\int_{-\infty}^{t}S^\dagger U_{21}\exp\left[-i\int_{-\infty}^{t'}\delta\,dt''\right]dt'\equiv\begin{bmatrix} D_2(t)\\ D_3(t)\end{bmatrix}\,,$$
$$|\,C_k(t)\,|^2=\left|\sum_{m=2}^{3}S_{km}D_m(t)\right|^2\,, \tag{6}$$

$$\delta(t)=-i\frac{dD^\dagger(t)}{dt}D(t)\exp\left[-2\int_{-\infty}^{t}\delta_I\,dt'\right]\,, \tag{7}$$

and

$$U_{21}=U_{12}^\dagger=\begin{bmatrix} u_{21}\\ 0\end{bmatrix}\,. \tag{8}$$

The function $D(t)$ is approximated by use of δ_r^0, which is the real part of δ, at time origin $t=0$ (the turning point of the nuclear motion), and superscript 0 means 0th approximation. The unitarity condition $D^\dagger(t)D(t)\leq 1$ is satisfied as follows. From (5) and (6) we have

$$D(t)=\int_{-\infty}^{t}S^\dagger U_{21}e^{-i\delta_r^0 t'}[1-D^\dagger(t')D(t')]^{1/2}dt'\,. \tag{9}$$

As the main contribution to the integral is supposed to come from the narrow region $t\simeq 0$ we put (see Sec. IV)

$$D(t)\approx[1-D^\dagger(t)D(t)]^{1/2}\int_{-\infty}^{t}S^\dagger U_{21}\exp(-i\delta_r^0 t')dt' \tag{10}$$

and lead

$$D(t)=(1+F^\dagger F)^{1/2}F\,,$$
$$F=\int_{-\infty}^{t}S^\dagger U_{21}\exp(-i\delta_r^0 t')dt'\,, \tag{11}$$

which obviously satisfies the unitarity constraint $D^\dagger(t)D(t)\leq 1$. Let us see the function δ_r^0, F, and the probability $|\,C_k\,|^2$ $(k=2,3)$, in more concrete forms. The phase δ_r^0, after simple procedure, is

$$\delta_r^0=\frac{|\,g_{21}^n\,|^2}{2\alpha v\rho^{2n-1}}\int_0^\infty[(\alpha-\beta)\sin(\Omega_1^0 x)$$
$$+(\alpha+\beta)\sin(\Omega_2^0 x)](1+x^2)^{-n/2}dx\,, \tag{12}$$

where $\Omega_1^0=(\rho/2v)\,|\,\Delta\omega-\alpha-\delta\omega\,|$ and $\Omega_2^0=(\rho/2v)\,|\,\Delta\omega+\alpha-\delta\omega\,|$. The exact quadrature (12) is unknown but is estimated by the approximation

$$S_n(A)=\int_0^\infty(1+x^2)^{-n/2}\sin(Ax)dx\simeq\frac{A}{n-2+A^2}\,, \tag{13}$$

which is a simple assumption from the both infinitely large and small limit of A (see Appendix) and an example, S_4, necessary in later sections, is illustrated in Fig. 2. We therefore have

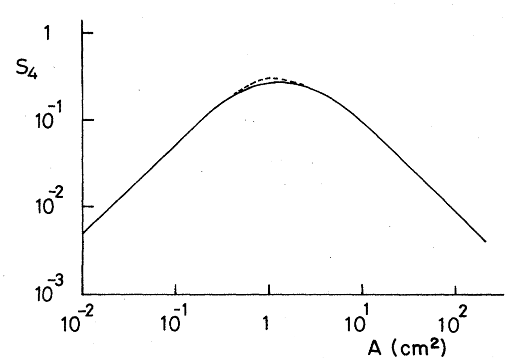

FIG. 2. The value of S_4 vs argument A. Solid line is the approximation (13) and broken line is the exact value.

$$\delta_r^0 = -\frac{|g_{21}^n|^2}{2\alpha\nu\rho^{2n-1}}\left[(\alpha-\beta)\frac{\Omega_1^0}{n-2+(\Omega_1^0)^2}+(\alpha+\beta)\frac{\Omega_2^0}{n-2+(\Omega_2^0)^2}\right], \tag{14}$$

$$F=\frac{g_{21}^n}{\nu\alpha\rho^{n-1}}\begin{bmatrix}(\alpha-\beta)Q_n(\Omega_1)K_{(n-1)/2}(\Omega_1)+(\alpha+\beta)Q_n(\Omega_2)K_{(n-1)/2}(\Omega_2)\\ 2u_{23}[Q_n(\Omega_1)K_{(n-1)/2}(\Omega_1)-Q_n(\Omega_2)K_{(n-1)/2}(\Omega_2)]\end{bmatrix}, \tag{15}$$

$$F^\dagger F=\left|\frac{g_{21}^n}{\nu\alpha\rho^{n-1}}\right|^2\{[(\alpha-\beta)^2+4|u_{23}|^2]Q_n^2(\Omega_1)K_{(n-1)/2}^2(\Omega_1)+[(\alpha+\beta)^2+4|u_{23}|^2]Q_n^2(\Omega_2)K_{(n-1)/2}^2(\Omega_2)\}, \tag{16}$$

where

$$Q_n(x)=\frac{\sqrt{\pi}(x/2)^{(n-1)/2}}{\Gamma(n/2)}, \tag{17}$$

$$|C_2|^2=\frac{1}{1+F^\dagger F}\left|\frac{g_{21}^n}{\nu\alpha\rho^{n-1}}\right|^2\{[(\alpha-\beta)Q_n(\Omega_1)K_{(n-1)/2}(\Omega_1)]^2+[(\alpha+\beta)Q_n(\Omega_2)K_{(n-1)/2}(\Omega_2)]^2\},$$

and (18)

$$|C_3|^2=\frac{1}{1+F^\dagger F}\left|\frac{g_{21}^n}{\nu\alpha\rho^{n-1}}\right|^2\{[Q_n(\Omega_1)K_{(n-1)/2}(\Omega_1)]^2+[Q_n(\Omega_2)K_{(n-1)/2}(\Omega_2)]^2\},$$

where

$$\Omega_1=\frac{\rho}{2\nu}|\Delta\omega-\alpha-\delta\omega-2\delta_r^0|,\quad \Omega_2=\frac{\rho}{2\nu}|\Delta\omega+\alpha-\delta\omega-2\delta_r^0|,$$

and $K_{(n-1)/2}$ is the modified Bessel function of $[(n-1)/2]$th order. The total cross section of the transition to the state $|f_3\rangle$ is

$$\sigma_3=2\pi\int_0^\infty \rho|C_3|^2 d\rho . \tag{19}$$

The argument Ω_2 in general is much larger than Ω_1 and thereby makes the function $K_{(n-1)/2}(\Omega_2)$ negligibly smaller than $K_{(n-1)/2}(\Omega_1)$ as the function decreases exponentially with argument. Hereafter, we disregard the term with Ω_2 and derive the dependence of σ_3 on the intensity of the power density.

A. Low power-density case

The argument Ω_1 in this case under the small detuning frequency is approximately given by

$$\Omega_1\approx\frac{|g_{21}^n|^2}{\nu\Delta\omega\rho^{2n-1}} . \tag{20}$$

The probability and cross section become

$$|C_3|^2=\frac{2^{3-n}\pi|u_{23}|^2}{\nu\Delta\omega\,\Gamma^2(n/2)}\Omega_1^{n-1/(2n-1)}\times K_{(n-1)/2}^2(\Omega_1)\left|\frac{g_{21}^n}{\nu\Delta\omega}\right|^{1/(2n-1)}, \tag{21}$$

$$\sigma_3=\frac{2^{4-n}|\pi u_{23}|^2}{(2n-1)\Gamma^2(n/2)}|g_{21}^n|^{6/(2n-1)}\times(\Delta\omega)^{2n/(2n-1)}\nu^{-(2n+2)/(2n-1)}A_n , \tag{22}$$

and

$$A_n=\int_0^\infty x^{n-(2n+1)/(2n-1)}K_{(n-1)/2}^2(x)dx . \tag{23}$$

The factor A_n is a constant dependent only on n and its value is listed in Table I for a few cases of n.

TABLE I. Example of A_n.

n	3	4	5	6
A_n	2.18	2.95	7.18	28.32

B. High power-density case

The phases δ_r^0, Ω_1, and $F^\dagger F$ are

$$\delta_r^0\simeq-\frac{(n-2)\Delta\omega|\nu g_{21}^n|^2}{\rho^{2n+2}|u_{23}|^4}, \tag{24}$$

$$\Omega_1\simeq\frac{\rho}{\nu}|\delta_r^0+|u_{23}||, \tag{25}$$

and

$$F^\dagger F\simeq 2^{4-n}\pi\left|\frac{u_{23}g_{21}^n}{\nu\alpha\rho^{n-1}\Gamma(n/2)}\right|^2\Omega_1^{n-1}K_{(n-1)/2}^2(\Omega_1) . \tag{26}$$

Direct analytical quadrature for the total cross section is unattainable but by looking minutely at Ω_1 and $F^\dagger F$ we can derive an intuitive formula. When the field is very strong the phase changes rapidly with an impact parameter ρ and $K_{(n-2)/2}(\Omega_1)$ vanishes sharply before and after the zero point of Ω_1 because a function $\Omega_1^2K_{(n-1)/2}^2(\Omega_1)$ decreases exponentially with the argument. The zero point ρ_0 of Ω_1 is

$$\rho_0=[(n-2)\Delta\omega\, v^2\,|\,g_{21}^n\,|\,]^{1/(2n+2)}\,|\,u_{23}\,|^{-5/(2n+2)}\ , \tag{27}$$

and is supposed to be very small and may cause a large maximum of $F^\dagger F$. As $\alpha-\beta$ is almost same with $2\,|\,u_{23}\,|$ and $F^\dagger F$ is much larger than unity at ρ_0 (and in its narrow vicinity), we obtain from (16) and (18) $|\,C_2\,|^2=|\,C_3\,|^2=0.5$. The total cross section σ_3 may be given by the simplest but fairly good formula, i.e.,

$$\sigma_3=2\pi\rho_0\,\Delta\rho_0\ , \tag{28}$$

where $\Delta\rho_0$ is half the width of $F^\dagger F$ to keep the significant probability and is decided as follows.

The function $F^\dagger F$ of (26) is rewritten as

$$F^\dagger F=2^{3-n}\pi^2\left|\frac{g_{21}^n}{v\rho^{n-1}\Gamma(n/2)}\right|^2 G_n \tag{29}$$

and

$$G_n(\Omega_1)=\begin{cases}1 \quad (n=2)\\ 1+\Omega_1 \quad (n=4)\\ 3+3\Omega_1+\Omega_1^2 \quad (n=6)\ ,\end{cases}$$

where the function G_n for odd n is an infinite series but the value is in between those of smaller and larger even numbers adjacent to n. The half-width $\Delta\rho_0$ is so defined as to keep $F^\dagger F>1$, i.e.,

$$2\left.\frac{d\Omega_1}{d\rho}\right|_{\rho=\rho_0}\Delta\rho_0\simeq[\ln(F^\dagger F)_{\rho=\rho_0}] \tag{30}$$

and then

$$\Delta\rho_0=\frac{v}{4(n+1)\,|\,u_{23}\,|}\times\left\{\ln[2^{3-n}\pi^2G_n(0)]+2\ln\left|\frac{g_{21}^n}{v\rho_0^{n-1}\Gamma(n/2)}\right|\right\}. \tag{31}$$

The total cross section (28) is written as

$$\sigma_3=\frac{\pi}{2(n+1)}\,|\,(n-2)\Delta\omega\, v^2 g_{21}^n\,|^{1/2(n+1)}\times v\,|\,u_{23}\,|^{-(2n+7)/(2n+2)}\times\ln\left\{2^{3-n}G_n(0)\left|\frac{\pi g_{21}^n}{v\rho_0^{n-1}\Gamma(n/2)}\right|^2\right\}. \tag{32}$$

The dependence of σ_3 on $|\,u_{23}\,|$ in the asymptotic region is

$$\sigma_3\propto|\,u_{23}\,|^{-(2n+7)/(2n+2)+\ln(\ln|u_{23}|)/\ln|u_{23}|} \tag{33}$$

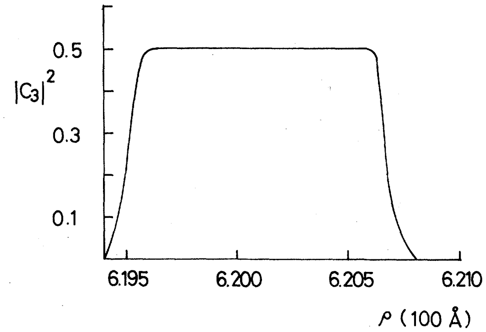

FIG. 3. Probability $|\,C_3\,|^2$ as the function of impact parameter at the relative velocity $v=10^3$ cm/sec and laser power density $P/A=10^{16}$ W/cm^2. The shape is almost square with its height of 0.5 and the width is nearly 0.0001 Å, which agrees with the value from (31).

and is $|\,u_{23}\,|^{-(n+3.5)/(n+1)}$ at $|\,u_{23}\,|$ limit.

III. THE CROSS SECTION OF THE PROCESS: $\mathrm{Sr}(5s5p\ ^1P_1)+\mathrm{Ca}(4s^2\ ^1S_0)+\hbar\omega \rightarrow \mathrm{Sr}(5s^2\ ^1S_0)+\mathrm{Ca}(3d4p\ ^1F_3)$

The process under consideration is diagrammed in Fig. 1 and the meaning of the notations are the same as those of Sec. II. Initially the energy is stored in the level $|\,a_2\rangle$ of Sr and Ca atoms is in the ground state $|\,b_1\rangle$. By collision, Sr makes a dipole transition to $|\,a_1\rangle$ and, on the contrary, Ca is excited to $|\,b_2\rangle$ via quadrupole transition, and goes to $|\,b_3\rangle$ by the dipole-laser interaction. Each parameter of Sec. II is replaced by ($n=4$ here)

$$g_{21}^n=\tfrac{3}{2}\mu_A q_B,\quad \mu_A=4\times10^{-18}\ \text{esu cm}\ ,$$
$$q_B=2.7\times10^{-26}\ \text{esu cm}^2\ ,$$
$$|\,u_{23}\,|=(2.6\times10^7\ \text{sec}^{-1/2})(P/A)^{1/2}\ ,$$

and

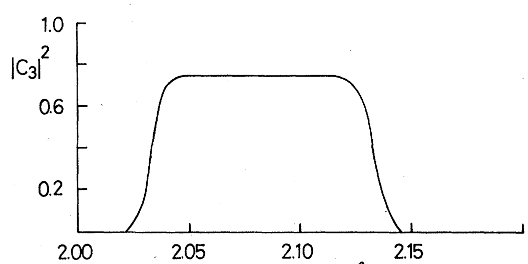

FIG. 4. Probability $|\,C_3\,|^2$ as the function of impact parameter at the relative velocity $v=10^3$ cm/sec and laser power density $P/A=10^{10}$ W/cm^2. The shape is nearly trapezoid with its height of 0.75 and the width is approximately 0.1 Å which agrees with the value from (31).

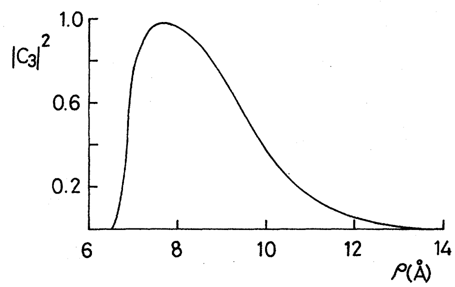

FIG. 5. Probability $|C_3|^2$ as the function of impact parameter at the relative velocity $v=10^3$ cm/sec and laser power density $P/A=10^8$ W/cm^2.

$$\Delta\omega=151\ \mathrm{cm}^{-1}\ .$$

All the parameters above are from Green *et al.*[15] except $|u_{23}|$ which is not clear and assumed as it is. The probabilities and total cross section calculated by (18) and (19) are shown in Figs. 3—6. When the field is weak, the probability and total cross section increase linearly with it as easily supposed and are confirmed experimentally. But the dependence on the dipole-quadrupole interaction is $|g_{21}^4|^{6/7}$ and $v^{-10/7}$ on the velocity. The behaviors in the intense field, to our knowledge, are unknown either theoretically or experimentally. We have shown intuitively that the cross section decreases in a way of (32), which follows well the curves of Fig. 6. Also shown are the transition probability vs impact parameter for two different intense-field cases. The latter case especially justifies the derivation of (28), i.e., the constant probability 0.5 in the narrow region of ρ and rapid decrease outside.

IV. DISCUSSION

We have investigated the dependence of the transition probability and cross section on the intensity of applied laser field. Also the general treatment is made for multipole interaction which causes the transition from the initial state to the virtual state. The cross section changes with $|g_{21}^n|^{6/(2n-1)}$, $v^{-(2n+5)/(2n+2)}$, and $|u_{23}|^2$ where the field is weak, that is to say, with the increase of multiplicity n, the dependence of σ_3 on g_{21}^n becomes weaker but stronger on velocity. The reason is understood that the interaction is dependent on R^{-n} and so the significant region becomes narrower with the n. In strong-field limit, σ_3 changes with $|u_{23}|^{-(2n+7)/(2n+2)}$, because the impact parameter ρ_0, which maximizes the probability, is proportional to $|u_{23}|^{-5/(2n+2)}$ and the width $\Delta\rho_0$ which is hardly dependent on n, changes with $|u_{23}|^{-1}$. Present theory guarantees the unitarity of probability by approximating (9) as (10). If the factor $[1-D^\dagger(t)D(t)]^{1/2}$ changes with time much more slowly than the other factor in the integrand of D and $D^\dagger$ the approximation is reasonable. To see this aspect we reconsider the (column) vector D

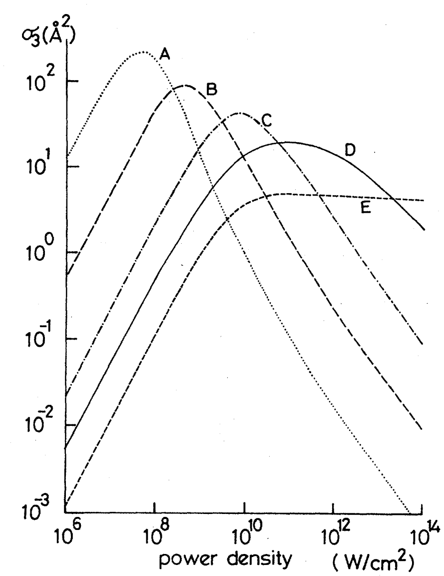

FIG. 6. Total cross section σ_3 vs laser power density at several relative velocities: *A*, $v=10^3$ cm/sec; *B*, $v=10^4$ cm/sec; *C*, $v=10^5$ cm/sec; *D*, $v=10^6$ cm/sec; *E*, $v=10^7$ cm/sec. The asymptotic behaviors of *A*, *B*, and *C* follow well to (33), but *D* and *E* are not yet in the asymptotic region.

$$D=\int_{-\infty}^{t} S^\dagger U_{21} e^{-i\delta_r^0 t'}[1-D^\dagger(t')D(t')]^{1/2}dt'$$
$$=\int_0^x W[1-D^\dagger(x')D(x')]^{1/2}dx'$$

where

$$x=\int_{-\infty}^{t}|u_{12}|dt'$$

and

$$W=\begin{bmatrix} S_{22}^* \\ -S_{32}\end{bmatrix}\exp\left[-\int_0^x \frac{\delta_r^0 dx'}{|u_{12}|}\right] .$$

Defined as

$$\gamma=\frac{\dot{W}^\dagger\dot{W}}{\left|\dfrac{d}{dx}[1-D^\dagger(x)D(x)]^{1/2}\right|^2} ,$$

(overdot denotes derivative by x) the ratio gives a criterion to the problem, namely if, in the region contributive to the integral, γ is much larger then unity, then the approximation is justified, but not otherwise. After straightforward algebra we have

$$\dot{W}=i(\Delta\omega-\delta_r^0)|u_{21}|W-i\begin{bmatrix} S_{32}^* \\ S_{22}\end{bmatrix}u_{32}\exp\left[i\int_0^x \frac{\Delta\omega-\delta_r^0}{|u_{12}|}dx'\right] ,$$

$$\dot{W}^\dagger\dot{W}=|u_{21}|^2[|u_{23}|^2+(\delta_r^0-\Delta\omega)^2] ,$$

$$\left|\frac{d}{dx}\sqrt{1-D^{\dagger}(x)D(x)}\right|^2 = |W^{\dagger}D+D^{\dagger}W|^2 = 4\left|\mathrm{Re}(S_{22}^{*}D_1 - S_{32}^{*}D_2)\exp\left[i(\Delta\omega-\delta_r^0)\int_0^x |u_{21}|^{-1}dx'\right]\right|^2$$
$$\leq 4(|S_{22}^{*}D_1|^2+|S_{32}^{*}D_2|^2) \leq 4(|D_1|+|D_2|)^2 \leq 8D^{\dagger}(x)D(x)$$

and then

$$\gamma \geq \frac{1}{8D^{\dagger}(x)D(x)}\left\{\left|\frac{u_{23}}{u_{21}}\right|^2+\left|\frac{\delta_r^0-\Delta\omega}{u_{21}}\right|^2\right\}.$$

The first two terms in the large parentheses above compensate each other, because when the laser field is weak and the first term is small, the second term becomes larger and vice versa. The ratio in actual process of Sec. III is 10^6 in the case of, say, Fig. 3. When the power density is low the factor $D^{\dagger}(x)D(x)$ is sufficiently small as known from (18) and (20) and makes γ large.

It is interesting to observe Green *et al.*'s result,[15] though correct comparison is impossible due to the different parametrization. Their experiment shows that the total cross section increases linearly with the weak-field intensity and seems to begin to saturate at about 10^{10} W/cm^2, although their surmise is the slow increase, even beyond it. Our example of $v=10^5$ cm/sec correspondent to their experiment shows saturation also at 10^{10} W/cm^2 and has the peak of 40 Å^2. The observed value at 7×10^9 W/cm^2 is several times larger than ours. The marked discrepancy between theory and experiment exist in the dependence of detuning frequency $\delta\omega$ on the line shape. High resolution experiments confirm the law of $\delta\omega^{-0.8}$ but theories on the contrary indicates $\delta\omega^{-0.5}$ in the case of dipole-dipole interaction. Our method in this respect is same with other theories. This discrepancy was supposed to come from the hypothesis of linear trajectory of nuclear motion.[16] Although the relative nuclear velocity is high where linear trajectory is pertinent our formulation does not reach to the experiment.

The comment above concerns the case of negative detuning frequency and the line shape on the side of positive $\delta\omega$ rapidly decreases with it. When a laser field is weak the total cross section satisfies[18]

$$\sigma_3 \propto \int_0^{\infty}\rho^{-3}z^2K_1^2(z)d\rho \rightarrow \delta\omega^{3/4}\exp(-A\,\delta\omega^{5/6}) \quad \text{as } \delta\omega\rightarrow\infty$$

where

$$z=\frac{\rho}{v}\left[\frac{(2\mu^{A_1}\mu^{B_1})^2}{3\Delta\omega\hbar^2\rho^6}+\delta\omega\right]$$

and

$$A=\frac{1}{6}\left[\frac{20(\mu^{A_1}\mu^{B_1})^2}{3v\Delta\omega\hbar}\right]^{1/6}$$

which except the factor of $\delta\omega^{3/4}$ agrees with Brechignac *et al.*'s analysis.[16]

Finally we wish to review the theory presented here. The results of the title process have characteristic features different from other studies[19] in that our cross section begins to decrease when the intensity of the laser field exceeds a certain point. Berman and Geltman[19] showed the increase of cross section with the intensity to the $1/(n-1)$ power ($n=3$ for dipole-dipole, $n=4$ for dipole-quadrupole). Origin of this difference comes from an intermediate state. The transition from $|f_1\rangle$ to $|f_3\rangle$ is made only via the virtual state $|f_2\rangle$. Initially, the Sr atom makes a dipole transition to ground state while the Ca atom is excited to the virtual level by quadrupole interaction. The Ca atom induced by an applied laser field then makes a dipole transition to the target state Ca($3d4p\ ^1F_3$) as is observed by Green *et al.*[15] The observation made by them indicates that the ratio of the rate of production of target-state Ca atoms by this prototype collision process is 450 times larger. Therefore the process is regarded as the prototype and the virtual state plays an indispensable role in it. If the prototype system is reduced forcibly to a two-state problem or the second perturbation is applied, the laser field acts as a direct coupling between the initial and final states. The cross section then keeps increasing with the intensity. So far as present theory is concerned, the cross section σ_3 decreases with it, the reason of which is as following. By the strong dipole-field interaction the atomic states $|b_2\rangle$ and $|b_3\rangle$ shift their energy by about $\mu^{B_2}E/2$ and, consequently, the energy difference between the first two states $|f_1\rangle$ and $|f_2\rangle$ also occurs only when both atoms approach close enough to each other to make the interaction large enough, i.e., the impact parameter to maximize the transition probability becomes smaller, with a more intense field, to cause a smaller cross section. The dependence of cross section on the relative velocity is common to general collision problems. When the velocity is low enough the system has enough time for complete transition from $|f_1\rangle$ to $|f_2\rangle$ on the way incoming and complete return to $|f_1\rangle$ on the way outgoing, and leaves no transition at all. High velocity, on the other hand, gives the system insufficient time for transition throughout the encounter. A velocity to maximize the probability is proportional to the interaction potential squared as is supposed from the Landau-Zener formula. As mentioned above, the dipole-quadrupole interaction is very large in the significant transition region when the field is intense. Thereby, the peak of cross shifts to the high-velocity side with the intensity.

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APPENDIX

We wish to derive an approximate relation (13). It is easily derived from the definition of $S_n(13)$ that when $A \ll 1$ we have

$$S_n \approx \int_0^\infty \frac{Ax}{(1+x^2)^{n/2}} dx = \frac{A}{n-2} \ . \tag{A1}$$

If n is even and is written as $n = 2m$ we have

$$S_{2m} = \int_0^\infty \sin(x)(A^2+x^2)^{-m} dx = A^{2m-1} \frac{(-1)^{m-1}}{(m-1)!} \frac{d^{m-1}}{dc^{m-1}} \left[\frac{1}{2A} \mathrm{Re}[e^{-A}E_1(-A-ix) - e^{A}E_1(A-ix)]_0^\infty \right] , \tag{A2}$$

where $C = A^2$ and E_1 is an exponential integral. The term in the square brackets in Eq. (A2) is written as

$$e^{-A}E_1(-A-ix) - e^{A}E_1(A-ix) \,|_0^\infty = \lim_{x\to\infty} \left[-\frac{e^{ix}}{A+ix} - \frac{e^{ix}}{A-ix} \right] - e^{-A}E_1(-A-i0) + e^{A}E_1(A-i0) \approx \frac{2}{A} \quad (A \gg 1) \ . \tag{A3}$$

Substituting (A3) into (A2) we have

$$S_n = A^{2m-1}/C^m = 1/A \ , \tag{A4}$$

which is independent of n. Combining (A1) and (A4), S_n of whole region of A is approximately given by

$$S_n = A/(n-2+A^2) \ . \tag{A5}$$

Although even n is assumed in deriving (A2), the function above may be interpolated to hold when n is odd.

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