New approach to the multistate problem and its application to the laser-induced transition process: $Sr(5s5p\,^1P^o)+Ca(4s^2\,^1S)+\hbar\omega \rightarrow Sr(5s^2\,^1S)+Ca(4p^2\,^1S)$

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We propose a new method to solve the multistate coupled equation. It overcomes the fatal defect of divergence which conventional perturbation theory has when the interaction is not small. We apply it successfully to laser-induced transition processes and obtain a total-cross-section formula.

I. INTRODUCTION

The Landau-Zener-Stuckelberg (LZS) formula[1] has been the most popular method for two-channel equations. However, the formula, in general, cannot treat the multichannel case nor give time-dependent approximate solutions.

In a previous paper[2] we succeeded in generalizing the LZS formula and broke through its difficulties by introducing a complex phase method. One of its successful points is that it can give a good probability amplitude at arbitrary time, which encourages us to go to the multichannel problem. Here "multi" means three or more.

In this paper a new approach to the multistate problem is proposed. In a previous paper we showed that the complex-phase method is superior to the perturbation theory in that it can take into account the effects of higher-order terms in a simple manner. The new idea is to (1) separate N states into $N-1$ and 1 states, (2) use the S matrix of $N-1$ states, and (3) regard the system as "two states" between $N-1$ and 1 states and use the complex-phase method. In other words, with the knowledge of $N-1$ states we can solve the case of N states. Of course, the two-states system is solved by use of our previous result. We apply this theory to the laser-induced transition process

$$\text{Sr}\ (5s5p\,^1P^o)+\text{Ca}\ (4s^2\,^1S)+\hbar\omega \rightarrow \text{Sr}\ (5s^2\,^1S)+\text{Ca}\ (4p^2\,^1S)\ ,$$

which may be simplified into a three-state model. The results are compared with those of other workers and show excellent improvement.[3–6]

II. THEORY

Theoretical study of the state-to-state transition in the N-state system is in general equivalent to solving the equation under the boundary condition $c_k(t=-\infty)=\delta_{k1}$:

$$\frac{d}{dt}\begin{bmatrix} c_1 \\ c_2 \\ \vdots \\ c_N \end{bmatrix} = i \begin{bmatrix} u_{11} & \cdots & u_{1N} \\ \vdots & & \vdots \\ u_{N1} & \cdots & u_{NN} \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ \vdots \\ c_N \end{bmatrix}, \quad u_{kk}=0\ , \tag{1}$$

where t is time, c_k is the probability amplitude for the "kth state" and u_{mn} is the matrix element between the mth and nth states [Eq. (1) is completely general and we need not specify the model or states yet].

On the assumption that the solution of $(N-1)$-state equations is available, i.e., that the following equation has been solved:

$$\frac{d}{dt}\begin{bmatrix} b_2 \\ b_3 \\ \vdots \\ b_N \end{bmatrix} = i \begin{bmatrix} u_{22} & \cdots & u_{2N} \\ \vdots & & \vdots \\ u_{N2} & \cdots & u_{NN} \end{bmatrix} \begin{bmatrix} b_2 \\ b_3 \\ \vdots \\ b_N \end{bmatrix} . \tag{2}$$

Then Eq. (1) becomes a "two-channel" problem,

$$\frac{d}{dt}\begin{bmatrix} C_1 \\ B \end{bmatrix} = i \begin{bmatrix} U_{11} & U_{12} \\ U_{21} & U_{22} \end{bmatrix} \begin{bmatrix} C_1 \\ B \end{bmatrix} , \tag{3}$$

where the column vector B and the elements of matrix U are

$$U_{11}=(U_{11}),\quad U_{12}=(u_{12}\ \ u_{13}\ \ \cdots u_{1N})\ ,$$

$$U_{21}=\begin{bmatrix} u_{21} \\ u_{31} \\ \vdots \\ u_{N1} \end{bmatrix},\quad U_{22}=\begin{bmatrix} u_{22} & \cdots & u_{2N} \\ \vdots & & \vdots \\ u_{N2} & \cdots & u_{NN} \end{bmatrix} ,$$

$$C_1=(c_1),\ B=\begin{bmatrix} c_2 \\ c_3 \\ \vdots \\ c_N \end{bmatrix} .$$

Hereafter we use the following symbols: asterisk, for complex conjugate; dagger, for Hermitian conjugate; superscript t (on symbols), for the transpose; and overdot, for the time derivative. When a unitary matrix S satisfies

$$\dot{S}=iU_{22}S \tag{4}$$

and such a vector Y is defined by

$$Y=S^{\dagger}B\ , \tag{5}$$

Eq. (3) becomes

$$\dot{C}_1 = iU_{12}SY ,$$
$$\dot{Y} = iS^{\dagger}U_{21}C_1 . \tag{6}$$

Since the matrix S satisfies the same equation as (2) we can use as S the scattering matrix of $N-1$ states already obtained and write it in the form

$$S = \begin{bmatrix} S_{22} & \cdots & S_{2N} \\ \vdots & & \vdots \\ S_{N2} & \cdots & S_{NN} \end{bmatrix} . \tag{7}$$

Eliminating Y from (7) we thereby have

$$\dot{C}_1 = -U_{12}S\int_{-\infty}^{t} S^{\dagger}U_{21}C_1 dt . \tag{8}$$

Though $U_{12}S$ is a vector as is known from (3), this integral equation is a scalar and is therefore the same as our previous result.[2] Thus the N-state problem is reduced to a single integral equation with no special approximation other than information on the $(N-1)$-state system. As the exact solution of this integral equation is beyond hope, we use the following complex-phase method, of which validity has been proved in a previous paper.

Rewriting C_1 by a complex phase δ as

$$C_1 = \exp\left[-i\int_{-\infty}^{t}\delta\, dt'\right] \tag{9}$$

and putting it into (9) we have the integral equation for δ:

$$\begin{aligned}\delta &= -i\exp\left[i\int_{-\infty}^{t}\delta\, dt'\right]U_{12}S \\ &\quad\times\int_{-\infty}^{t}S^{\dagger}U_{21}\exp\left[-i\int_{-\infty}^{t'}\delta\, dt''\right]dt' \\ &= -i\exp\left[-2\int_{-\infty}^{t}\delta_I dt'\right]\dot{D}^{\dagger}(t)D(t) ,\end{aligned} \tag{10}$$

where

$$D \equiv \int_{-\infty}^{t} S^{\dagger}U_{21}\exp\left(-i\int_{-\infty}^{t'}\delta\, dt''\right)dt'$$

and δ_I is the imaginary part of δ.

The procedure above is only the transformation of argument from C_1 to δ and no new improvement seems to have been made; however, important progress has been made as will be seen later in actual use. The amplitude and probability of the first state are then

$$C_1 = \exp\left[-i\int_{-\infty}^{t}\dot{D}^{\dagger}(t')D(t')\exp\left[-2\int_{-\infty}^{t'}\delta_I dt''\right]dt'\right] \tag{11}$$

and the other amplitudes are given from (5), (7), and (10) as

$$\begin{bmatrix} c_2 \\ c_3 \\ \vdots \\ c_N \end{bmatrix} = iSD(t) . \tag{12}$$

Let us examine the unitarity of the probability. Rewriting (10) as

$$2\delta_I = \exp\left[-2\int_{-\infty}^{t}\delta_I dt'\right]\{\dot{D}^{\dagger}(t)D(t) + [\dot{D}^{\dagger}(t)D(t)]^*\} \tag{13}$$

and recalling the relation

$$[\dot{D}^{\dagger}(t)D(t)]^* = \dot{D}^{t}(t)D^*(t) = [D^{\dagger}(t)\dot{D}(t)]^{t} \tag{14}$$

and that $D^{\dagger}(t)\dot{D}(t)$ is a scalar we have

$$2\delta_I\exp\left[2\int_{-\infty}^{t}\delta_I dt'\right] = -\frac{d}{dt}[D^{\dagger}(t)D(t)] . \tag{15}$$

By quadrature of (15) under the boundary condition $c_1(-\infty)=1$ we have

$$|c_1|^2 = \exp\left[2\int_{-\infty}^{t}\delta_I dt'\right] = 1 - D^{\dagger}(t)D(t) \tag{16}$$

or, by use of (12) and (16), the unitarity $\sum_{k=1}^{N}|c_k|^2 = 1$.

The transition probability to every other k state is given from (12) by

$$|c_k|^2 = \left|\sum_{m=2}^{N} S_{km}D_m(t)\right|^2 , \tag{17}$$

where $D_m(t)$ is the mth element of the vector $D(t)$. So far we have derived the exact relations and solutions for the problem and to go further some approximations must be made. The lowest-order approximation for $D(t)$ is given by neglecting all in the right-hand side of it:

$$D(t) \approx D^0(t) = \int_{-\infty}^{t} S^{\dagger}U_{21}dt' \tag{18}$$

which, via (16) and (18), leads to the results of the perturbation theory.

It is obvious that those $|c_k|^2$ are not necessarily within unity and often exceed it. From the definition of $D(t)$ it is understood that what guarantees unitarity is δ, which suggests that adequate choice of δ is of primary importance in this theory. The next-order approximation is based on the use of the first-order complex phase and the corresponding $D^1(t)$:

$$D^1(t) \equiv \int_{-\infty}^{t} S^{\dagger}U_{21}\exp\left[-i\int_{-\infty}^{t'}\delta^0 dt''\right]dt$$

with

$$\delta^0 = -iU_{12}S\int_{-\infty}^{t} S^{\dagger}U_{21}dt' \tag{19}$$

or from (10), (11), and (18)

$$\begin{aligned}D^1(t) &= \int_{-\infty}^{t} S^{\dagger}U_{21}e^{-D^{0\dagger}(t')D^0(t')/2} \\ &\quad\times\exp\left[-i\int_{-\infty}^{t'}\delta_R^0 dt''\right]dt'\end{aligned} \tag{20}$$

where δ_R^0 is the real part of δ^0.

If the main contribution to the integral (20) comes from the narrow region of $t \cong 0$ and $[D^0(t)]^{\dagger}D^0(t)$ is a slowly varying function there we can simplify $(D^1)^{\dagger}D^1$ more as

$$\begin{aligned}[D^1(\infty)]^{\dagger}D^1(\infty) &= [D^2(\infty)]^{\dagger}D^2(\infty) \\ &\quad\times\exp\{-[D^0(0)]^{\dagger}D^0(0)\} ,\end{aligned} \tag{21}$$

where

$$D^2(\infty) = \int_{-\infty}^{\infty} S^{\dagger}U_{21}e^{-i\delta_R^0 t}dt$$

which improves the probability much better. The important conclusion of this section is that if the solution of the $(N-1)$-state system is available we can solve the N-state problem approximately.

III. APPLICATION TO THE LASER-INDUCED TRANSITION PROCESS: $\mathrm{Sr}(5p5s\ ^1P^o)+\mathrm{Ca}(4s^2\ ^1S)+\hbar\omega \rightarrow \mathrm{Sr}(5s^2\ ^1S)+\mathrm{Ca}(4p^2\ ^1S)$

We apply the theory developed in the preceding section to the title process. Since Harris and Lidow[4] attempted to make the first observation of a laser-stimulated energy transfer of this process, some theoretical works for it have been published. All of the methods, however, are based on the perturbation theory. Harris and White[5] started with a three-state problem and reduced it into a two-state problem and then solved the coupled equation numerically. Geltman[6] derived the formula of the cross section by use of the second perturbation theory. Knight[3] also used the perturbation theory and solved the coupled equation under the assumption that the matrix element is slowly varying compared with the energy difference and derived the formula of the cross section.

What is common to all the formulas derived theoretically so far is that the probability diverges at a small impact parameter limit (linear trajectory has been assumed), because the dipole-dipole interaction has a singularity at zero nuclear distance. This difficulty is inevitable so long as the perturbation theory is used.

In this section we will show that our theory can avoid the difficulty and our formulas of the probability and cross section work well. As our theoretical model of the process is taken from Harris and co-worker we do not repeat their explanation except when it is necessary.

The system is simplified to have three pertinent states and the wave function is expanded in a set of product eigenfunctions of infinitely separated atoms:

$$\Psi=\sum_{n=1}^{3} c_n\,|f_n\rangle e^{-i\omega_n t}\,, \tag{22}$$

where $\omega_n=E_n/\hbar$, $|f_1\rangle=|a_2\rangle|b_1\rangle$, $|f_2\rangle=|a_1\rangle|b_2\rangle$, $|f_3\rangle=|a_1\rangle|b_3\rangle$, and the states $|a_n\rangle,|b_n\rangle$ and energy E_n are illustrated in Fig. 1. The interaction Hamiltonian is

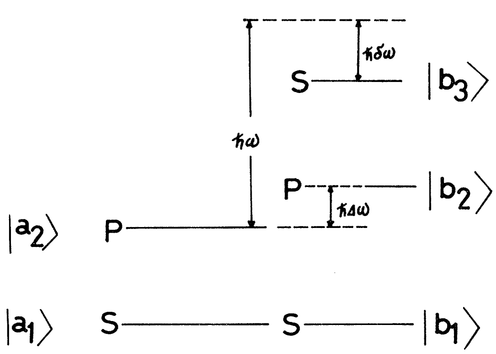

FIG. 1. Schematic diagram of colliding atom A and B.

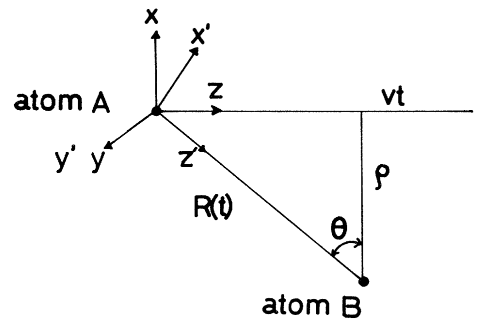

FIG. 2. Coordinate of the system. Straight line is assumed for the trajectory of atom A. Primed coordinate system is space fixed and unprimed one is body fixed. They are related to each other by the angle θ.

$$H=(Y_A-Y_B)eE\cos(\omega t)+(e^2/R^3)(X'_AX'_B+Y'_AY'_B-2Z'_AZ'_B)\,, \tag{23}$$

where $E\cos(\omega t)$ is the laser field with frequency ω, R is the relative nuclear distance, and the coordinates are shown in Fig. 2.

Linear trajectory is assumed for R: $R=(\rho^2+v^2t^2)^{1/2}$, where ρ is an impact parameter and v is relative velocity. The coupled equations are

$$\frac{d}{dt}\begin{bmatrix}c_1\\c_2\\c_3\end{bmatrix}=i\begin{bmatrix}0 & u_{12} & 0\\ u_{21} & 0 & u_{23}\\ 0 & u_{32} & 0\end{bmatrix}\begin{bmatrix}c_1\\c_2\\c_3\end{bmatrix}, \tag{24}$$

where

$$u_{12}=\frac{2\mu^{A_1}\mu^{B_1}}{\sqrt{3}\hbar R^3}e^{-i\Delta\omega t},\quad u_{21}=u_{12}^{*}\,,$$

$$u_{23}=\frac{\mu^{B_2}E}{2\hbar}e^{i(\Delta\omega+\delta\omega)t},\quad \mu^{A_1}=\langle a_2\,|\,ez_A\,|\,a_1\rangle\,,$$

$$\mu^{B_1}=\langle b_1\,|\,eZ_B\,|\,b_2\rangle,\quad \mu^{B_2}=\langle b_2\,|\,eY_B\,|\,b_3\rangle\,,$$

and $\delta\omega$ is the detuning frequency.

The S matrix in this case is decided by the two-channel solution of the states $|f_2\rangle$ and $|f_3\rangle$ as

$$S=\begin{bmatrix}S_{22} & -S_{32}^{*}\\ S_{32} & S_{22}^{*}\end{bmatrix}, \tag{25}$$

where

$$S_{22}=\frac{e^{i\beta t/2}}{2\alpha}[(\alpha-\beta)e^{i\alpha t/2}+(\alpha+\beta)e^{-i\alpha t/2}]\,,$$

$$S_{32}=\frac{|u_{23}|}{\alpha\hbar}e^{-i\beta t/2}(e^{i\alpha t/2}-e^{-i\alpha t/2})\,,$$

with $\alpha=[(\Delta\omega+\delta\omega)^2+4|u_{23}|^2]^{1/2}$ and $\beta=\Delta\omega+\delta\omega$.

Recalling the definition of U_{nm} in (3), we have the matrix elements of the case:

$$U_{21}=\begin{bmatrix}u_{21}\\0\end{bmatrix},\quad U_{12}=U_{21}^{\dagger},\quad U_{22}=\begin{bmatrix}0 & u_{23}\\ u_{32} & 0\end{bmatrix}. \tag{26}$$

As the dipole-dipole interaction $|u_{12}|$ has a maximum at $t=0$, the transition is expected mostly in the narrow region centered by the point and therefore formula (21) is applicable now.

The expression of $D^2(\infty)$ is derived by substituting (24)—(26) into (21):

$$D^2(\infty)=\frac{\mu^{A_1}\mu^{B_1}}{\sqrt{3}\alpha v\hbar\rho^2}\int_{-\infty}^{\infty}(1+x^2)^{-3/2}e^{-(i\rho/v)\delta_R^0 x}\begin{bmatrix}(\alpha-\beta)e^{i\rho(\Delta\omega-\alpha-\delta\omega)x/2v}+(\alpha+\beta)e^{i\rho(\Delta\omega+\alpha-\delta\omega)x/2v}\\ -\dfrac{2E\mu^{B_2}}{\hbar}(e^{i\rho(\Delta\omega+\alpha-\delta\omega)x/2v}-e^{-i\rho(\Delta\omega-\alpha-\delta\omega)x/2v})\end{bmatrix}dx\ . \tag{27}$$

It follows from (19) and (24) that

$$\delta^0(0)=-\frac{4i(\mu^{A_1}\mu^{B_1})^2}{3\hbar^2\rho^3}\int_{-\infty}^{0}\frac{S_{22}^{*}e^{i\Delta\omega t dt}}{R^3}dt=-\frac{2i\mu^{A_1}\mu^{B_1}}{\sqrt{3}\hbar\rho^3}(1,0)D^0(0)\ . \tag{28}$$

The necessary parameters are

$$\frac{4(\mu^{A_1}\mu^{B_1})^2}{3\hbar^2\Delta\omega}=4.2\times10^7\ \mathrm{cm^{-1}\,\mathring{A}^6},\quad \frac{(E\mu^{B_2})^2}{4\hbar^2\Delta\omega}=5.5\times10^{-10}\ \mathrm{cm^{-1}(P/A)\,W/cm^2},\quad \mathrm{P/A}=5\times10^5\ \mathrm{W/cm^2}\ . \tag{29}$$

The quantities $D^2(\infty)$ and $D^0(0)$ are therefore

$$D^2(\infty)=\frac{2\mu^{A_1}\mu^{B_1}}{\sqrt{3}v\hbar\rho^2\alpha}\begin{bmatrix}(\alpha-\beta)ZK_1(Z)+(\alpha+\beta)Z'K_1(Z')\\ E\dfrac{\mu^{B_2}}{\hbar}[ZK_1(Z)-Z'K_1(Z')]\end{bmatrix},$$

$$D^0(0)=-\frac{i\mu^{A_1}\mu^{B_1}}{\sqrt{3}v\alpha\hbar\rho^2}\begin{bmatrix}\dfrac{\alpha-\beta}{1+\Omega_1^2}\Omega_1+\dfrac{\alpha+\beta}{1+\Omega_2^2}\Omega_2\\ \dfrac{E\mu^{B_2}/\hbar}{1+\Omega_1^2}\Omega_1-\dfrac{E\mu^{B_2}/\hbar}{1+\Omega_2^2}\Omega_2\end{bmatrix}+\tfrac{1}{2}D^0(\infty)\ , \tag{30}$$

where K_1 is the modified Bessel function,

$$\Omega_1=\frac{\rho}{2v}(\Delta\omega-\alpha-\delta\omega),\quad \Omega_2=\frac{\rho}{2v}(\Delta\omega+\alpha-\delta\omega)\ ,$$

$$Z=\Omega_1-\frac{\rho}{v}\delta_R^0(0),\quad Z'=\Omega_2-\frac{\rho}{v}\delta_R^0(0)\ ,$$

and we have used the approximation

$$\int_0^{\infty}\frac{\sin(Ax)}{(1+x^2)^{3/2}}dx\approx\frac{A}{1+A^2} \tag{31}$$

the validity of which is understood in Fig. 3.

In this model it holds that

$$\frac{\alpha-\beta}{\alpha}\approx\frac{(E\mu^{B_2})^2}{2\alpha^2}\ll1 \tag{32}$$

and except a very small impact parameter ρ; the frequency Ω_2 is so large that $\Omega_2K_2(\Omega_2)$, which decreases exponentially with Ω_2, becomes negligible to lead to

$$D^2(\infty)=-\frac{\mu^{A_1}\mu^{B_1}E\mu^{B_2}}{\sqrt{3}\alpha v\hbar^2\rho^2}ZK_1(Z)\begin{bmatrix}0\\1\end{bmatrix},$$

$$D^0(0)=-\frac{i\mu^{A_1}\mu^{B_1}}{\sqrt{3}\alpha\rho^3\hbar}\begin{bmatrix}1\\0\end{bmatrix}+\tfrac{1}{2}D_0(\infty)\ , \tag{33}$$

where we have used

$$\frac{(\alpha+\beta)\Omega_2}{1+\Omega_2^2}\approx\frac{2v}{\rho}\ . \tag{34}$$

It follows thereby from (21) that

$$D^1(\infty)=-\frac{2\mu^{A_1}\mu^{B_1}E\mu^{B_2}}{\sqrt{3}v\alpha\hbar^2\rho^2}ZK_1(Z)\begin{bmatrix}0\\1\end{bmatrix}\times e^{-[D^0(0)]^{\dagger}D^0(0)/2} \tag{35}$$

with

$$Z=\Omega_1-\delta_R^0(0)\frac{\rho}{v}=\frac{\rho}{v}\left[\frac{4(\mu^{A_1}\mu^{B_1})^2}{3\Delta\omega\hbar^2\rho^6}-\delta\omega\right].$$

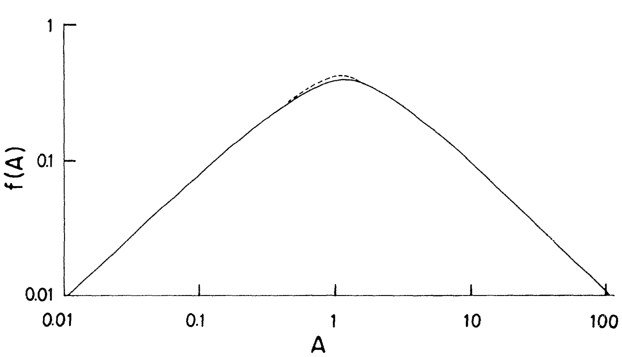

FIG. 3. Value of $f(A)=\int_0^{\infty}(1+x^2)^{-3/2}\sin(Ax)dx$ as the function of parameter A. Solid line is the approximation $A/(1+A^2)$ and broken line is the exact value.

From (21) and (35) we have

$$[D^1(\infty)]^\dagger D^1(\infty)=\left[\frac{\mu^{A_1}\mu^{B_1}E\mu^{B_2}}{\sqrt{3}\,\Delta\omega\, v\hbar^2\rho^2}\right]^2 Z^2K_1^2(Z)\exp\left[-\left[\frac{\mu^{A_1}\mu^{B_1}E\mu^{B_2}}{\sqrt{3}v\hbar^2\Delta\omega\rho^2}\Omega_1K_1(\Omega_1)\right]^2\right]. \tag{36}$$

The transition probability $|c_2|^2$ is calculated from (17) and (25) and is given by

$$|c_2(\infty)|^2=\left|S_{22}(\infty)\int_{-\infty}^{\infty}u_{12}^*S_{22}^*\exp\left[-i\int_{-\infty}^{t}\delta^0dt'\right]dt+S_{32}^*(\infty)\int_{-\infty}^{\infty}U_{12}^*S_{32}\exp\left[-i\int_{-\infty}^{t}\delta^0dt'\right]dt\right|^2. \tag{37}$$

By the same reason discussed to derive (33) the first term of (37) is extremely small and should be neglected. The second term is also negligible because it is the order of $|u_{12}u_{23}^2|$ which is far smaller than $[D^1(\infty)]^\dagger D^1(\infty)$ in this model.

When the probability is small enough as it is now the exponential function of (35) is replaced by unity and therefore the final probability is simplified as

$$|c_3(\infty)|^2=\left[\frac{\mu^{A_1}\mu^{B_1}E\mu^{B_2}}{\sqrt{3}\Delta\omega\hbar^2v\rho^2}\right]^2 Z^2K_1^2(Z)\,, \tag{38}$$

where we have used the second equation of (13) and the discussion that $|c_2(\infty)|^2$ is negligible.

The total cross section σ is given by

$$\sigma=2\pi\int_0^\infty \rho\,|c_3|^2d\rho\,. \tag{39}$$

When the detuning frequency is zero we can simplify more:

$$\sigma=\frac{2}{5}\pi\left[\frac{4(\mu^{A_1}\mu^{B_1})^2}{3\hbar^2\Delta\omega v}\right]^{3/5}\frac{(E\mu^{B_2})^2}{4\hbar^2\Delta\omega}\times\int_0^\infty Z^{7/5}K_1^2(Z)dZ$$

$$=0.87\pi\left[\frac{4(\mu^{A_1}\mu^{B_1})^2}{3\hbar^2\Delta\omega}\right]^{3/5}\frac{(E\mu^{B_2})^2}{4\hbar^2\Delta\omega}v^{-8/5} \tag{40}$$

which is the same formula given by Harris and co-worker if the factor of 0.87 is replaced with their 5.9.

IV. DISCUSSION

We have developed the theory to give approximate solution of the N-state coupled equation by the S matrix of $N-1$ states and applied it to the three-state problem of the laser-induced transition process. We wish to view how the theory works in the discussion of the calculation.

Several authors presented formulas of this laser-induced process which is reduced to three-state model though Harris and Lidow considered five levels. Knight's work is based on the second perturbation theory and shows the divergence of probability with decreasing impact parameter. Therefore, he introduced a "cutoff" procedure to cover it. This cutoff procedure introduced artificially by physical insight has strong influence over the results, namely the transition probability and cross section are strongly dependent on the size of the boundary impact parameter. However, the divergence difficulty should be removed in the theory so long as the physical model is reasonable. While conventional perturbation theory does not work, the present theory has broken through it by the complex phase method.

What our theory insists is that the complex phase in the integrand of $D(t)$ suppress the divergence of interaction without fail by rapidly changing the real phase

$$\exp\left[\frac{-i\rho}{v}\int_{-\infty}^{t}\delta_R dt'\right]$$

and decreasing the imaginary phase

$$\exp\left[\frac{\rho}{v}\int_{-\infty}^{t}\delta_I dt'\right].$$

Harris and White's theory is based on the intuitive abbreviation that intermediate state $|f_2\rangle$ is sufficiently small throughout the process and deformed the three-state coupled equation into that of two states. Although the effects of the intermediate state are partly included in their theory an inconsistency occurs, namely the unitarity holds only between the first and third state, which causes a larger transition probability.

Figure 4 is the transition probability versus impact parameter in the case of $\delta\omega=0$ and is about $\frac{1}{10}$ smaller than theirs, though both curves behave similarly and have the maxima at the same impact parameter.

Let us see the behavior of the probability P as the function of ρ. The upper limit of P is known from (36) and (38) by

$$P=|c_3(\infty)|^2\le\left[\frac{\mu^{A_1}\mu^{B_1}E\mu^{B_2}}{\sqrt{3}\hbar^2\Delta\omega v\rho^2}\right]^2=\frac{0.94}{\rho^4}$$

which is smaller than theirs by about $\frac{1}{10}$. The total cross section versus detuning frequency is shown in Fig. 5. The difference of the total cross section is, of course, due to

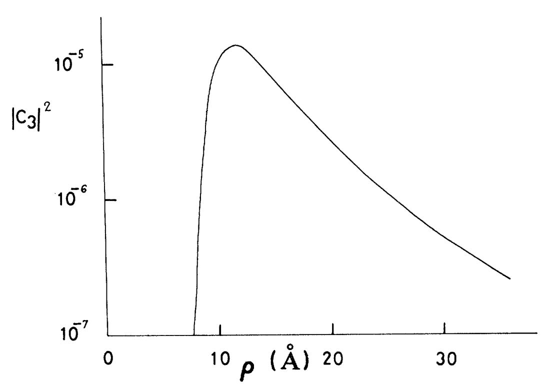

FIG. 4. Transition probability vs impact parameter.

that of the probability, i.e., the present result is a factor of nearly $\frac{1}{10}$ smaller. The cross section increases linearly with the strength of laser field when the transition is small but may saturate and decrease again beyond a certain strength, because of the exponential function in the definition of $D^1(\infty)$. Our formula (40) is surprisingly the same with theirs except it is a factor 1/6.8 smaller.

This model is unsuitable to the LZS formula because it requires a narrow transition region near the energy crossing point as well as enough separation between other transition regions. As all the energy separations have no zero point and the matrix element $|u_{23}|$ is assumed constant in the entire region no transition region can be defined at least between the second and third states. Therefore, there is no room for the LZS formula in this process.

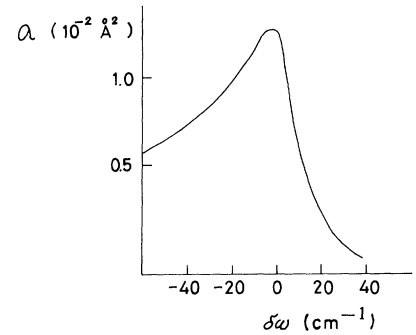

FIG. 5. Dependence of the total cross section on the detuning laser frequency (wave number).

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