# Modification of the far-wing absorption profile due to collisional coherence

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A phenomenon occurring in a near-resonant collision between two dissimilar atoms in the presence of a laser field is described. Owing to a collision-induced coherence, the adiabatic collisional dressed states are found to behave as superradiant and subradiant Dicke states, giving rise to a marked asymmetry in the far wings of the absorption line shape.

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## I. INTRODUCTION

In laser spectroscopy, when an atom is driven to a coherent superposition of states, a quantum-mechanical interference between transition amplitudes takes place, leading to narrow features in absorption and emission line shapes. In most processes of atomic physics, coherences are created by laser fields, occasionally in combination with electric or magnetic fields, and damped by collisional processes. In the past few years, the study of atomic collisions in the presence of radiation fields has shown that the collisional interaction can play a predominant role in the creation of atomic coherences. Collisionally aided excitation of a coherence between electronic levels has been found responsible for the so-called pressure-induced extra resonances (PIER's) appearing in four-wave-mixing generation [1] as well as in fluorescence emission [2,3].

In this paper we present a theoretical study of collisionally aided radiative excitation (CARE) showing that, under appropriate circumstances, the absorption profile in the line wings is significantly affected by the presence of collision-induced coherences. Following the first discussion by Lisitsa and Yakovlenko in 1974 [4], the CARE process, also referred to as "optical collision," has been extensively studied, both theoretically and experimentally, for an atom interacting with a structureless perturber [5–7]. In this case, the absorption spectrum is characterized by an asymmetric shape showing, for an attractive dipole-dipole potential, an extended red wing, where the laser detuning from resonance $\Delta=\omega_L-\omega_0$ is compensated by the collisional shift of atomic levels. In this region, defined as static wing, for $-\Delta\gg 1/\tau_c$, where $\tau_c$ is a typical collision time, the absorption process can be described, following the Born-Oppenheimer approximation, as an instantaneous transition between adiabatic quasimolecular states. The absorption cross section in the static wing follows the power law $\sigma(\Delta)\propto|\Delta|^{-3/2}$, typical of a pure van der Waals wing [8].

In the present paper we are interested in a near-resonant CARE, when the colliding atoms, represented as two-level systems, have near-resonant excited levels. The energy defect between the excited levels $2\delta$ is supposed large enough ($2\delta\gg 1/\tau_c$) to make the collision adiabatic in the absence of laser field. During a short-range collision, when the interaction energy is of the order of $2\delta$, the system of dissimilar atoms behaves as a system of similar atoms. The transition from van der Waals to resonance interaction, studied theoretically by Nikitin [9], manifests as a double-slope behavior for the absorption cross section in the static wing $\sigma(\Delta)\propto|\Delta|^{-3/2}(|\Delta|+2\delta)^{-1/2}$. An observation of this effect was reported by Niemax for the europium-strontium system [10].

It is important to emphasize that, in previous treatments of near-resonant CARE, the radiative coupling of the perturber atom was neglected. In our approach, where we consider both colliding atoms $A$ and $B$ coupled to the radiation field with $n$ photons of frequency $\omega_L$, the absorption reaction can follow either path

$$A+B+n\hbar\omega\rightarrow\begin{cases}A^*+B+(n-1)\hbar\omega_L\\ A+B^*+(n-1)\hbar\omega_L\end{cases}, \tag{1}$$

where $A^*$ and $B^*$ denote atomic excited states.

Although the relative weight of each path in reaction (1) depends on the associated dipole moment and on the laser detuning, a strong collisional coupling may allow two transition paths leading to the same final state. For instance, the excitation of atom $A$ can take place directly by absorption of a photon or indirectly by a collisional energy transfer following a photon absorption by atom $B$. The quantum interference between these transition amplitudes can produce a large modification of the far-wing absorption profile. It could be expected that in a cell experiment, owing to the velocity distribution of colliding partners and to the collisional statistics, any effect of col-

 

lisional coherences would be washed out. It is an important result of the present study to show that, under conditions to be satisfied in experiments, the collision-induced interference effect takes place, giving rise to a frequency-selective transparency in the absorption spectrum of the transient molecule. The basic interference is interpreted with reference to the superradiant and subradiant Dicke states, introduced to describe the collective behavior of the quasimolecular system. In this framework the induced collision interference effect is equivalent to the coherent population trapping phenomenon, which produces narrow features in the laser spectroscopy of three-level systems [11]. Cooperative transitions of a quasimolecular system have also been considered by Lam and Rand in a treatment of stimulated pair emission, showing a decoupling of the static dipole-dipole interaction induced by a strong laser field of proper frequency [12]. Superradiant and subradiant molecular states have been conveniently used for the description of collisions among ultracoldlike atoms in optical traps by Julienne [13]. The collective behavior of two identical atoms coupled by dipole-dipole interaction has been proposed by Knight and coworkers as a possible explanation for the observed correlation in quantum jumps of trapped ions [14].

As it will be shown in the last part of the paper, the present interference effect is quite similar to that occurring in a three-level atomic system driven by two near-resonant laser fields. In that case, the collisionally aided excitation of atomic states, dressed by the laser fields, leads to the pressure-induced extra resonances in atomic populations [3].

## II. PHYSICAL PICTURE

Consider a near-resonant collision between two dissimilar atoms in the presence of a laser field. The atoms are assumed to be two-level quantum systems, as shown in Fig. 1. We will study the radiative transitions of the colliding atoms stimulated by the laser field (absorption or emission), following a methodology that has proved to be very convenient for the treatment of laser-assisted collisions [15]. The compound system $A$-$B$ is described in the basis of the relevant product states

$$|0\rangle=|\alpha\rangle|\beta\rangle;\quad |1\rangle=|\alpha^*\rangle|\beta\rangle;\quad |2\rangle=|\alpha\rangle|\beta^*\rangle \tag{2}$$

with the respective energies $E_0=0$, $E_1=\hbar\omega_0$, $E_2=\hbar(\omega_0-2\delta)$, having introduced the energy defect $2\delta=[E(\alpha^*)-E(\beta^*)]/\hbar$. For the collisional interaction we assume the long-range dipole-dipole potential in the scalar form

$$V(t)=\frac{V_0}{R^3(t)} . \tag{3}$$

During a collision with impact parameter $b$ and relative speed $v$, under the assumption of classical, rectilinear trajectories, the internuclear separation $R$ varies as $\sqrt{b^2+v^2t^2}$. If the energy defect is large compared to $\tau_c^{-1}$, where $\tau_c$ is the typical duration of a collision, the collision can be considered adiabatic.

The amplitude of the linearly polarized electromagnetic field is written as

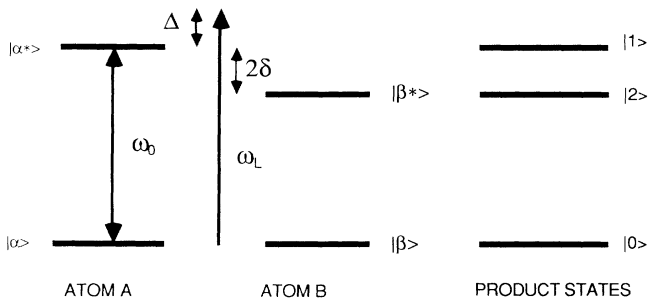


FIG. 1. Configuration of energy levels assumed for the colliding atoms. The relevant product states are shown on the right.

$$E=\frac{E_0}{2}(e^{i\omega_L t}+e^{-i\omega_L t}) , \tag{4}$$

where $E_0$ is assumed constant during the collision. The strength of the atom-field interaction is given by

$$\Omega_A=-E_0\mu_A/2\hbar,\quad \Omega_E=-E_0\mu_B/2\hbar , \tag{5}$$

where $\mu_A$ and $\mu_B$ are the transition dipole moments of atoms $A$ and $B$, respectively. The laser detuning is measured through $\Delta=\omega_L-\omega_0$, the frequency offset from the transition $|0\rangle-|1\rangle$, and through $\Delta'=-\Delta-2\delta$, the frequency offset from the transition $|0\rangle-|2\rangle$. If both laser detunings from the atomic transition frequencies $|\Delta|$ and $|\Delta'|$ are larger than $\tau_c^{-1}$, transitions can take place only during the collision, owing to the van der Waals shift of atomic levels. In this case, since the collision duration ($\tau_c\approx 1$ps) is typically much shorter than the radiative lifetime ($\tau_r\approx 10$ns), we can neglect spontaneous emission in the Hamiltonian evolution of the quasimolecular states.

Following a standard dressed-state picture, we introduce the atom+field states $|0,n\rangle$, $|1,n-1\rangle$, $|2,n-1\rangle$ of energy $E_{0,n}=\hbar\Delta$, $E_{1,n-1}=0$, and $E_{2,n-1}=-2\delta\hbar$, respectively. By expanding the wave function of the compound system in this basis

$$|\Psi\rangle=a_0|0,n\rangle+a_1|1,n-1\rangle+a_2|2,n-1\rangle , \tag{6}$$

we have the Schrödinger equation

$$i\dot{\bar{a}}=H\bar{a},\quad \bar{a}=\begin{bmatrix} a_0 \\ a_1 \\ a_2 \end{bmatrix} \tag{7}$$

with the Hamiltonian

$$H=\begin{bmatrix} \Delta & \Omega_A & \Omega_B \\ \Omega_A & 0 & V \\ \Omega_B & V & -2\delta \end{bmatrix} . \tag{8}$$

By performing the unitary transformation

$$\bar{b}=T\bar{a} \tag{9}$$

with

$$T=\begin{bmatrix}1 & 0 & 0\\ 0 & \cos\theta & \sin\theta\\ 0 & -\sin\theta & \cos\theta\end{bmatrix}, \tag{10}$$

the equations of motion are transformed into

$$i\dot{\bar{b}}=(H'+\dot{T}T^{-1})\bar{b}\ , \tag{11}$$

where

$$\bar{b}=\begin{bmatrix}b_0\\ b_1\\ b_2\end{bmatrix},\quad H'=THT^{-1}\ . \tag{12}$$

For $\tan2\theta=V/\delta$ the collisional interaction can be diagonalized and the transformed Hamiltonian results in

$$H'=\begin{bmatrix}\Delta & \Omega_a\cos\theta+\Omega_b\sin\theta & -\Omega_a\sin\theta+\Omega_b\cos\theta\\ \Omega_a\cos\theta+\Omega_b\sin\theta & \lambda_+ & 0\\ -\Omega_a\sin\theta+\Omega_b\cos\theta & 0 & \lambda_-\end{bmatrix}. \tag{13}$$

Here

$$\lambda_+=-\delta+\sqrt{\delta^2+V^2},\quad \lambda_-=-\delta-\sqrt{\delta^2+V^2} \tag{14}$$

are eigenvalues corresponding to the adiabatic collisional dressed states

$$|+,n-1\rangle=\cos\theta|1,n-1\rangle+\sin\theta|2,n-1\rangle\ ,$$
$$|-,n-1\rangle=-\sin\theta|1,n-1\rangle+\cos\theta|2,n-1\rangle\ . \tag{15}$$

In the limit $V\to0$ the eigenvalues $\lambda_+$ and $\lambda_-$ approach $E_1$ and $E_2$, respectively. If the condition $|\dot{\theta}|\ll|\lambda_+-\lambda_-|$, implying

$$\left|\frac{\dot{V}}{V}\right|\approx\frac{1}{\tau_c}\ll2\delta \tag{16}$$

is satisfied, then the collisional interaction can be treated adiabatically, and the matrix $\dot{T}T^{-1}$ can be neglected in the equation of motion (11), which can be written as

$$i\dot{\bar{b}}=H'\bar{b}\ . \tag{17}$$

The schematic time dependence of the eigenvalues (14) during the collision for laser detunings $\Delta>0$ and $\Delta<-2\delta$ ($\Delta'>0$) are shown in Figs. 2(a) and (b), respectively.

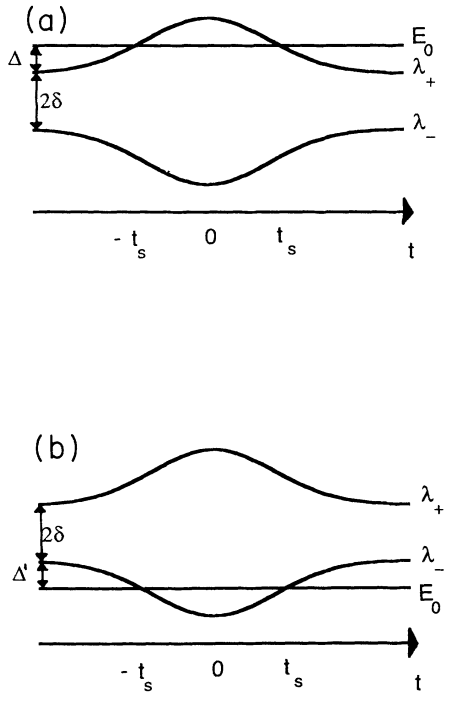


FIG. 2. Schematic variation of the energy levels of adiabatic dressed states during a collision for (a) laser detuning on the blue side of state $|1\rangle$ (blue wing); (b) laser detuning on the red side of state $|2\rangle$ (red wing). According to the quasistatic approximation, instantaneous transitions occur at the stationary-phase points $t=\pm t_s$.

## III. PERTURBATION SOLUTION

Equation (17) can be solved through a perturbation treatment in the radiation-atom interaction, since the laser field, acting only as a probe of the collisional interaction, will be assumed very weak. We limit ourselves to considering the absorption alone, assuming both atoms are initially in their ground states,

$$b_0(-\infty)=a_0(-\infty)=1\ ,$$
$$b_+(-\infty)=a_1(-\infty)=0\ , \tag{18}$$
$$b_-(-\infty)=a_2(-\infty)=0\ .$$

To first order in $\Omega_A$, $\Omega_B$ for the dressed- state amplitudes, we find the following solutions:

$$b_0(t)=\exp(-i\Delta t)\ ,$$
$$b_+(t)=-i\exp\left[-i\int_{-\infty}^{t}\lambda_+ d\tau\right]\int_{-\infty}^{t}(\Omega_A\cos\theta+\Omega_B\sin\theta)\exp\left[-i\int_{-\infty}^{t'}(\Delta-\lambda_+)dt''\right]dt'\ , \tag{19}$$
$$b(t)=-i\exp\left[-i\int_{-\infty}^{t}\lambda_- d\tau\right]\int_{-\infty}^{t}(\Omega_A\sin\theta-\Omega_B\cos\theta)\exp\left[-i\int_{-\infty}^{t'}(\Delta-\lambda_-)dt''\right]dt'\ .$$

If $\Delta>0$ or $\Delta<-2\delta$, the major contribution to the integrals in (19) comes from the points of stationary phase in the integrands. As in Ref. [15] two points of stationary phase ($\pm t_s$) exist for a sufficiently small impact parameter (see Fig. 2). For $|\Delta|\gg\tau_c^{-1}$ in the first case and $|\Delta+2\delta|\gg\tau_c^{-1}$ in the second case, the stationary-phase approximation can be used [16], yielding for the transition probabilities

$$|b_+(+\infty)|^2\approx\frac{4\pi}{|\ddot{\phi}_1(t_s)|}|\Omega_A\cos\theta_s+\Omega_B\sin\theta_s|^2\ ,$$
$$|b(+\infty)|^2\approx\frac{4\pi}{|\ddot{\phi}_2(t_s)|}|\Omega_A\sin\theta_s-\Omega_B\cos\theta_s|^2\ , \tag{20}$$

where

$$\phi_\pm(t)=\int_{-\infty}^{t}[\Delta-\lambda_\pm(t')]dt' \tag{21}$$

and $t_s$ is the positive solution of either equation,

$$\Delta-\lambda_\pm(t_s)=0\ . \tag{22}$$

In the derivation of (20) we have neglected, following Ref. [15], the rapidly oscillating contribution to the final-state amplitude due to the interference between the transition amplitude at $t=-t_S$ and $t=t_S$.

From the relations

$$R_s=\sqrt{b^2+v^2t_s^2}\ ,\ \sin\theta_s=\frac{1}{\sqrt{2}}\left[\frac{\Delta}{\Delta+\delta}\right]^{1/2}\ ,$$
$$\cos\theta_s=\frac{1}{\sqrt{2}}\left[\frac{\Delta+2\delta}{\Delta+\delta}\right]^{1/2}\ , \tag{23}$$

we find, for a laser detuning $\Delta>0$ [Fig. 2(a)],

$$|b_+(+\infty)|^2=\frac{2\pi V_0}{3v}\frac{[\Omega_A(\Delta+2\delta)^{1/2}+\Omega_B\Delta^{1/2}]^2}{\Delta^{3/2}(\Delta+2\delta)^{3/2}}\times\frac{1}{R_s^2[1-(\frac{b}{R_s})^2]^{1/2}}\ , \tag{24}$$

and since in this case $\Delta-\lambda_-\geq 2\delta$, $|b_-(+\infty)|^2\approx 0$.

By integrating over the impact parameter distribution and averaging over the Maxwellian distribution of interatomic speed, we get the following absorption cross section in the blue wing:

$$\sigma_B\approx 2\pi\int_0^\infty f(v)dv\int_0^{R_s}|b_+(+\infty)|^2 b\,db$$
$$=\frac{16\pi V_0}{3\bar{v}}\frac{[\Omega_A(\Delta+2\delta)^{1/2}+\Omega_B\Delta^{1/2}]^2}{\Delta^{3/2}(\Delta+2\delta)^{3/2}}\ , \tag{25}$$

where $\bar{v}$ is the mean interatomic speed at the cell temperature $T$.

On the other hand, for a laser detuning $\Delta<-2\delta$ [Fig. 2 (b)], it is the integral providing $b_-$ which may have stationary-phase points when $\lambda_-=-\Delta-2\delta$, in which case $\Delta-\lambda_+\approx 2\delta$. Following the same procedure as before, we get, for the absorption cross section in the red wing versus the detuning $\Delta'$, the following expression:

$$\sigma_R\approx\frac{16\pi V_0}{3\bar{v}}\frac{[\Omega_A\Delta'^{1/2}-\Omega_B(\Delta'+2\delta)^{1/2}]^2}{\Delta'^{3/2}(\Delta'+2\delta)^{3/2}}\ . \tag{26}$$

Equations (25) and (26) show that the far-wing absorption profiles for laser detunings on the blue side of state $|1\rangle$

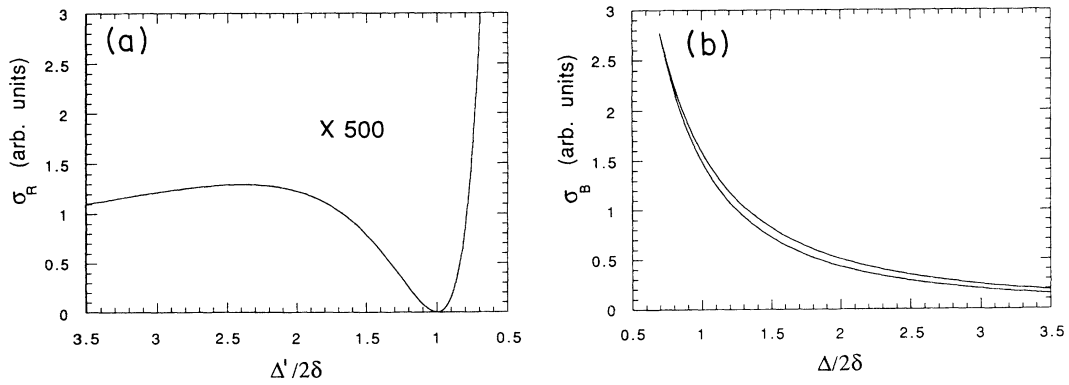


FIG. 3. Behavior of the absorption cross section vs the normalized detuning in the red wing (a) and in the blue wing (b). The strength of the radiative couplings of atoms $A$ and $B$ are in the ratio $\Omega_A/\Omega_B=1/\sqrt{2}$. The lower curve (b) represents the double-slope behavior obtained neglecting the radiative coupling of the perturber atom ($\Omega_B=0$) multiplied by the normalization factor 3.64.

(blue wing $\Delta>0$) and on the red side of state $|2\rangle$ (red wing $\Delta<-2\delta$) can be different, although $\lambda_+(V)$ and $\lambda_-(V)$ have symmetrical behavior. Neglecting the coupling of the perturber atom with the radiation field, namely $\Omega_B$ in Eq. (25) and $\Omega_A$ in Eq. (26), the absorption profile follows in both wings the double-slope law, typical of a near-resonant CARE process [10,17],

$$\sigma_B(\Delta)\alpha\Delta^{-3/2}(\Delta+2\delta)^{-1/2}\ ,$$
$$\sigma_R(\Delta')\alpha(\Delta')^{-3/2}(\Delta'+2\delta)^{-1/2}\ . \tag{27}$$

As presented above, the change of the slope from $-\frac{3}{2}$, for $\Delta,\ \Delta'\ll 2\delta$, to $-2$ for $\Delta,\ \Delta'\gg 2\delta$ is evidence of the transition from a van der Waals to resonant interaction. This behavior has been checked, for instance, by measurements of the fluorescence emitted by europium atoms perturbed by collisions with strontium atoms [10]. The effect of the radiative coupling of the perturber atom on the absorption spectrum may be noticed in Figs. 3(a) and 3(b), where both the red- and blue-wing profiles given by Eqs. (25) and (26) are reported for $\Omega_A/\Omega_B=\sqrt{2}$. In Fig. 3(b) we have also reported the absorption profile of the simple CARE process when the coupling of the perturber atom with the laser field is neglected. It may be noticed that in the blue wing the radiative coupling of the perturber has only a minor influence on the absorption profile. On the contrary, this effect can be dramatic in the case of Eq. (26) (red wing), describing a strongly depressed wing. Moreover, the red wing, for $\Omega_A>\Omega_B$ exhibits a transparency point, in correspondence to a detuning satisfying the condition

$$\frac{\Omega_A}{\Omega_B}=\frac{\cos\theta_s}{\sin\theta_s}=\left[\frac{\Delta'+2\delta}{\Delta'}\right]^{1/2}\ . \tag{28}$$

The significant change of the red-wing profile was not observed in the europium-strontium experiment of Ref. [10], where the attention was reserved to the blue-wing profile. Owing to the minor change produced by the perturber, observation of the blue-wing profile by itself does not provide a clear evidence of the radiative coupling of the perturber.

## IV. DISCUSSION AND CONCLUSION

An interpretation of this effect can be easily given in the dressed-state picture, considering the case of a detuning large compared to the energy defect. In fact, for $|\Delta|\gg 2\delta$, from Eq. (23) it results in $\theta_s\to\pi/4$. Consequently the collisional dressed states at the stationary-phase points approach the symmetric and antisymmetric superpositions of bare states,

$$|+,n-1\rangle\to(1/\sqrt{2})|1,n-1\rangle+(1/\sqrt{2})|2,n-1\rangle\ , \tag{29}$$
$$|-,n-1\rangle\to-(1/\sqrt{2})|1,n-1\rangle+(1/\sqrt{2})|2,n-1\rangle\ .$$

Assuming for simplicity $\Omega_A=\Omega_B$, Eqs. (25) and (26) reduce to

$$\sigma_B(\Delta)\approx\frac{16\pi V_0}{3\bar{v}}\frac{4\Omega_A^2}{\Delta^2}\ ,\quad \sigma_R(\Delta')\approx 0\ . \tag{30}$$

Since the blue wing is due to a transition towards the $|+,n-1\rangle$ state, while the red wing is due to a transition towards the $|-,n-1\rangle$ state, in the limiting case of a resonant collision the collisional dressed states behave as collective Dicke states: the upper dressed state behaves as a superradiant state, with a coupling strength $2\Omega_A$ towards the $|0,n\rangle$ state, while the lower dressed state behaves as a subradiant state, being not coupled to the $|0,n\rangle$ state.

In the uncoupled-state basis, the difference between the blue- and the red-wing profiles can be interpreted in terms of quantum-mechanical interference between two transition pathways. As shown in the diagrammatic representation of Fig. 4(a), state $|2\rangle$ can be excited directly by collision-induced absorption of a photon. In addition, during a close collision, owing to the strong coupling between states $|1\rangle$ and $|2\rangle$, there is a possibility of excitation of state $|1\rangle$ by collision-induced absorption of a photon, followed by collisional transfer to state $|2\rangle$ [Fig. 4(b)]. In the case of strong collisional interaction, these transition amplitudes appear at the same order of perturbation theory, giving rise to an interference effect. This description shows a close similarity to that given by Grynberg and Berman for PIER in the dynamics of a three-level atom driven by two laser fields [3]. The main differences between the PIER process and that investigated here are the following. (i) In the case of PIER, states $|1\rangle$ and $|2\rangle$ belong to an atomic system and are coupled by a two-photon transition, while in the case of CARE they belong to a transient molecule and are coupled by an interatomic potential. (ii) The laser detuning, in the case of PIER, should be in the impact region ($|\Delta|\ll\tau_c^{-1}$), while in the case of CARE it should be in the static region ($|\Delta|\gg\tau_c^{-1}$).

In conclusion, we have presented a theoretical study of a near-resonant optical collision showing that the far-wing absorption profile can be significantly affected by optical coherences created by the interatomic potential, despite the collisional statistics. In fact, we have shown that the interference effect in the absorption profile is not

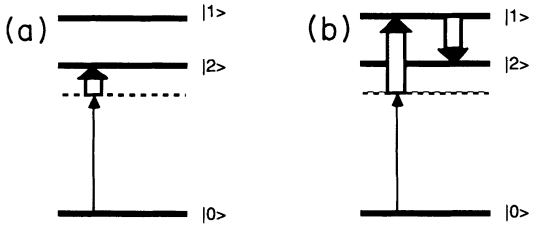


FIG. 4. Diagrammatic representation of the collisionally aided excitation of state $|2\rangle$ in the bare-state picture. (a) Direct collision-induced transition to state $|2\rangle$; (b) pathway involving a collision-induced transition to state $|1\rangle$ followed by collisional excitation transfer to state $|2\rangle$.

washed out by (i) dependence of the potential on the interatomic separation, (ii) spread of atomic velocities and impact parameters in the collisions. Since, in our approach, both atoms are considered coupled to the radiation field, the distinction between an active and perturber atom in the single collision is not significant. In an actual experiment, the relative active and perturber roles can be defined only by the relative densities of the two species, which should be optimized in order to depress the influence of collisions between similar atoms. In effect, in order to evidence the phenomenon treated in this work, the measurement of the blue-wing profile requires traces of species $A$ and a higher density of species $B$, as done in the europium-strontium experiment by Niemax [10]. The opposite case is required for the observation of the red wing. These conditions, together with the problems caused by dimer absorption, have probably prevented this effect from being brought into evidence in the many experiments performed on collisions between different alkali-metal atoms.

The approach presented in this paper concerns collisions of two atomic species $A$ and $B$ with different absorption spectra in the presence of a laser field. In the case of resonant collisions the treatment is very similar, and the main conclusion remains the same: An asymmetry in the absorption profile is expected with a strongly depressed red wing. Once again we should ask whether or not this phenomenon could be observed or has been observed experimentally. It has to be pointed out that for alkali-metal atoms, where most observations of absorption line profile have been made, the presence of several hyperfine levels and several crossings or anticrossings in the quasimolecular potential curves certainly mask this phenomenon. For this reason, we believe that the experimental investigation of the effect should be concentrated on other atomic species.

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