Collisional decoherence in the presence of ultrafast optical pulses

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An expression for the van der Waals interaction between an active atom and a perturber atom is derived using an irreducible tensor representation. This interaction leads to magnetic state decoherence in an ensemble of active atoms. The magnetic state decoherence can be suppressed by the application of ultrafast optical pulses. It is shown that the depolarization cross section can be suppressed by as much as 40% with only four optical pulses during a single collision and that, for a large number of pulses, the cross section is proportional to $n_0^{-2/11}$ where n_0 is the number of pulses at the Weisskopf radius. This paper generalizes our previous results in which the collisional interaction was modeled as a square pulse.

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Several recent papers [1–3] have contained proposals for suppressing the decoherence resulting from the coupling of a quantum system to a thermal reservoir. The methods discussed in these papers require the application of time-dependent perturbations to the system on a time scale that is short compared to the correlation time of the reservoir. In this paper we apply a similar idea to magnetic state relaxation in an atomic vapor.

''Active'' atoms in a thermal vapor interacting with a bath of foreign gas perturbers undergo elastic collisions that cause a depolarization of any initial magnetic state coherence present in the active atoms. More precisely, if we consider active atoms that are initially prepared in the $m=0$ sublevel of a $J=1$ state, then collisions with the perturbers will cause the population to equilibrate among the $m=0,\pm 1$ sublevels at a rate Γ_{col} that is typically on the order of 10^7-10^8 s^{-1} per Torr of perturber pressure.

The depolarization arises from elastic collisions that can be described by a semiclassical model in which the perturbers move along classical trajectories. It is common to model the atom-perturber interaction as a van der Waals interaction varying as $R(t)^{-6}$ where the quantity $R(t)$ is the distance between the active atom and perturber. The collision time τ_c is approximately equal to b/v where b is the impact parameter and v is the relative speed of the two atoms. For thermal speeds, and an impact parameter b_0 equal to the so-called Weisskopf radius that serves as a characteristic radius in collisional decoherence, τ_c is of order 1 ps.

To suppress the magnetic state decoherence one must perturb the active atoms on a time scale much less than τ_c in order to disrupt the coherent evolution of the atomic state wave function resulting from the van der Waals interaction. This necessitates the use of interactions that have durations on the order of 10–100 fs. One can achieve such interactions using ultrafast optical pulses applied to the initial states of the active atoms.

The modification of collisional dynamics in the presence of intense laser fields, i.e., $\chi\tau_c \gtrsim 1$, where χ is the Rabi frequency, is not a new subject. See, for example, the papers [5–8] and the review [9]. However, all theoretical treatments, including [7], which specifically treats time-dependent fields, have considered the field strength to be constant over the course of a collision so that the dressed states of the atom+field system were the appropriate basis states for studying the collision dynamics. However, we wish to turn the field on and off several times during the course of a single collision. Consequently, the previous work is inapplicable here.

The experiment that we envision involves an active atom initially in a $J_a=0$ ground state, $|g,J_a=0,m=0\rangle$, at $t=0$, that is excited by a pulse of duration $\tau_e \ll \tau_c$ to the $m=0$ sublevel of a $J_a=1$ excited state, $|e,J_a=1,m=0\rangle$. It is assumed that the spontaneous emission decay rate of the excited state is much smaller than Γ_{col} so that spontaneous decay on the time scale of magnetic state decoherence can be ignored. A series of off-resonant linearly polarized ultrafast pulses with duration $\tau_p \ll \tau_c$ is applied that couple the states $|e,1,m\rangle$ to a $J_a=0$ state, $|u,0,0\rangle$. This is depicted in Fig. 1. When the atom-field detuning δ on the e-u transition is large compared with τ_p^{-1}, the net effect of each pulse is to produce a phase shift $\Delta_s\tau_p$ of the $|e,1,m\rangle$ state amplitudes. Explicitly, one finds that this ac Stark effect phase shift is given by $\Delta_s\tau_p=\tau_p|\chi_{e1m,u00}|^2/\delta$, where $\chi_{e1m,u00}$ is a Rabi frequency associated with the e-u transition that is proportional to the amplitude of the external field pulse. The field strengths are chosen such that the phase shift of the $m=0$ state amplitude due to each of the pulses is a uniformly distributed random number in the interval $[0,2\pi]$. This type of

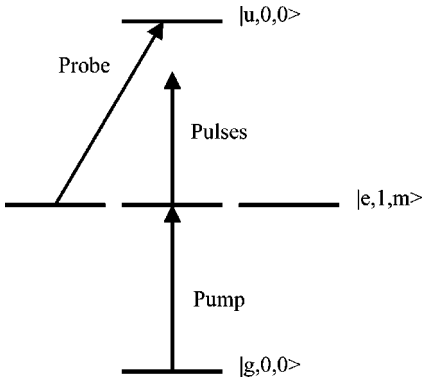

FIG. 1. Atomic level diagram illustrating proposed experiment. The pump and probe fields are resonant on the $|g,0,0\rangle \rightarrow |e,1,0\rangle$ and $|e,1,1\rangle \rightarrow |u,0,0\rangle$ transitions, respectively. The pulses are detuned from the $|e,1,0\rangle \rightarrow |u,0,0\rangle$ transition by an amount δ.

pulse sequence could be achieved using pulse shaping techniques [10]. The effect of the pulses is to disrupt the coherent collision process in an irreversible manner owing to the stochastic nature of the phases $\Delta_s \tau_p$. The pulse train must be on continuously for a time of order Γ_{col}^{-1}. After a time Γ_{col}^{-1}, the population in the $m=\pm 1$ states is probed with a circularly polarized pulse.

In a previous paper [4] we modeled the collision using a square pulse and neglected the collisional level shifts of the Zeeman states. In this paper we wish to consider a van der Waals interaction calculated from first principles. The resulting interaction is not isotropic and depends on the relative orientation of the atom and perturber. The average over collision orientations becomes nontrivial in the presence of directional external fields. The direct approach would be to numerically solve the density-matrix equations for a suitably large enough set of collision geometries to obtain an accurate estimate of the spatially averaged cross sections. Instead, we look for a ''typical collision'' that gives the same cross section for a particular transition in the perturbation theory limit and in the absence of any external field, and use this geometry to calculate the cross section when an external field is present.

I. INTERACTION POTENTIAL

The dipole-dipole interaction between two colliding atoms can be written as [11]

$$V(t)=\sum_{Q,K}\left[-\frac{(4\pi)^{3/2}}{R(t)^3}\left(\frac{2}{15}\right)^{1/2}Y^*_{2,Q}(\theta(t),\phi(t))\delta_{K,2}\right]$$

$$\times\sum_{q,q'}\langle 1,q;1,q'|2,Q\rangle T^1_q(a)T^1_{q'}(p), \tag{1}$$

where $T^k_q(a)$ and $T^k_q(p)$ are the electric multipole moment operators of the active (a) and perturber (p) atoms in an irreducible basis, and the coupling coefficient is a Clebsch-Gordan coefficient. If the active atom is located at the origin while the perturber moves along the trajectory $\mathbf{R}(t)=x(t)\hat{\mathbf{x}}+y(t)\hat{\mathbf{y}}+z(t)\hat{\mathbf{z}}$, then $\cos\theta(t)=z(t)/R(t)$, $\cos\phi(t)=x(t)/\sqrt{x(t)^2+y(t)^2}$.

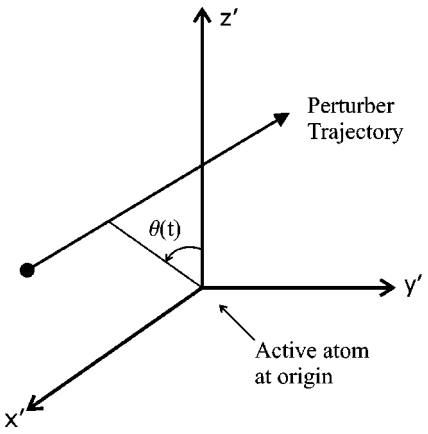

FIG. 2. The ''standard collision geometry'' in which the perturber moves along the trajectory $\mathbf{R}(t)=vt\hat{\mathbf{x}}'+b\hat{\mathbf{z}}'$ in the $x'z'$ plane.

For straight line trajectories in the laboratory frame, $\mathbf{R}(t)=\mathbf{v}t$, one can define a collision frame as depicted in Fig. 2. In the collision frame, the perturber moves along the ''standard'' trajectory $\mathbf{R}(t)=vt\hat{\mathbf{x}}'+b\hat{\mathbf{z}}'$. The collision frame is related to the laboratory frame by a rotation. Consequently, the average over all collision orientations corresponds to an average over the three Euler angles that parametrize the rotation to the collision frame.

In the case of nonresonant foreign gas perturbers, both atoms undergo virtual transitions to allowed excited states, but return to their initial angular-momentum multiplet if the collision is adiabatic. Adiabaticity is satisfied if $\tau_c^{-1}\ll\omega_{pa}$ where ω_{pa} is the frequency difference or sum between the virtual transitions of the active and perturber atoms. In this case, for an active atom having total angular momentum $J_a=1$ and perturber atom having ground-state angular momentum $J_p=0$, one finds that the state amplitudes of the active atom evolve as

$$i\dot{a}_m=\sum_{m'}U_{mm'}(t)a_{m'}, \tag{2}$$

where

$$U_{mm'}(t)=\frac{1}{\hbar^2\omega_{pa}}\sum_{m''}\langle 1,m;0,0|V(t)|0,0;1,m''\rangle\langle 0,0;1,m''|V(t)|1,m';0,0\rangle$$

$$=\sum_{m''}\sum_{Q,N,q,q',n,n'}\left[\frac{1}{\hbar^2\omega_{pa}}\frac{(4\pi)^3}{R(t)^6}\left(\frac{2}{15}\right)Y^*_{2,Q}(\theta(t),\phi(t))Y^*_{2,N}(\theta(t),\phi(t))\right]|T|^2$$

$$\times\langle 1,q;1,q'|2,Q\rangle\langle 1,n;1,n'|2,N\rangle\langle 0,0;1,q|1,m\rangle\langle 1,m'';1,q'|00\rangle\langle 1,m';1,n|0,0\rangle\langle 0,0;1,n'|1,m''\rangle$$

$$=\sum_{m'',Q,Q'}(-1)^{Q'}\frac{|C|^2}{R(t)^6}Y^*_{2,Q}(\theta(t),\phi(t))Y^*_{2,Q'}(\theta(t),\phi(t))\langle 1,m;1,-m''|2,Q\rangle\langle 1,-m';1,m''|2,Q'\rangle, \tag{3a}$$

$$|T|^2=\frac{1}{9}|\langle J=1||T^{(1)}(a)||J=0\rangle|^2|\langle J=1||T^{(1)}(p)||J=0\rangle|^2, \tag{3b}$$

$$|C|^2=\left(\frac{2}{15}\right)\frac{(4\pi)^3}{\hbar^2\omega_{pa}}|T|^2, \tag{3c}$$

and $\langle J=1||T^{(1)}(a,p)||J=0\rangle$ is a reduced matrix element.

Even though the above result was derived for the specific case of the perturber making a $J_p=0\rightarrow J_p=1$ virtual transition while the active atom made a $J_a=1\rightarrow J_a=0$ virtual transition, the above result is valid when all allowed virtual transitions are included [12]. Only the definition of $|C|^2$ is changed. Rewriting the spherical harmonics using the composition rule for spherical harmonics,

$$Y^*_{2,Q}(\theta,\phi)Y^*_{2,Q'}(\theta,\phi)=\sum_{L,l}\sqrt{\frac{25}{4\pi(2L+1)}}\langle 2,Q;2,Q'|L,l\rangle \times\langle 2,0;2,0|L,0\rangle Y^*_{L,l}(\theta,\phi),$$

one then obtains after some manipulations the final form for the matrix elements

$$U_{mm'}(t)=\sum_{L,l}(-1)^{m'+L+1}\frac{25|C|^2}{\sqrt{4\pi(2L+1)}R(t)^6} \times\langle 1,m;1,-m'|L,l\rangle\langle 2,0;2,0|L,0\rangle \times\begin{Bmatrix}1&1&L\\2&2&1\end{Bmatrix}Y^*_{L,l}(\theta(t),\phi(t)), \tag{4}$$

where $\{\}$ is a Wigner 6-J symbol. The 6-J symbol is zero unless $0\leqslant L\leqslant 2$ and $\langle a,0;b,0|c,0\rangle=0$ if $a+b+c=$ odd integer.

It follows that $U_{mm'}(t)$ contains only spherical harmonics of order $L=0,2$. For an arbitrary collision geometry that is related to the standard geometry by *active* rotations through the angles α,β,ξ about the z, y, and z axes, respectively, one finds

$$U_{mm'}(t,\alpha,\beta,\xi)=\sum_{L,l}(-1)^{m'+L+1}\frac{25|C|^2}{\sqrt{4\pi(2L+1)}R(t)^6} \times\langle 1,m;1,-m'|L,l\rangle\langle 2,0;2,0|L,0\rangle \times\begin{Bmatrix}1&1&L\\2&2&1\end{Bmatrix} \times D^{(L)}_{ll'}(\alpha,\beta,\xi)Y^*_{L,l'}(\theta_0(t),\phi_0(t)). \tag{5}$$

Here $\tan\theta_0(t)=vt/b$ and $\phi_0(t)=0$ in the collision frame. In the remainder of this paper we focus on the $m=0\rightarrow m=1$ transition, with associated matrix element

$$U_{01}(t,\alpha,\beta,\xi)=[U_{10}(t,\alpha,\beta,\xi)]^* =\frac{-|C|^2}{R(t)^6}\frac{5}{4}\sqrt{\frac{1}{15\pi}}\sum_m D^{(2)}_{-1m}(\alpha,\beta,\xi) \times Y^*_{2,m}(\theta_0(t),\phi_0(t)). \tag{6}$$

II. INCLUSION OF EXTERNAL PULSES

The external pulses can be treated using an impact approximation provided that there is no significant change in the expectation values of the active-atom variables during the pulse duration. This corresponds to the conditions

$$\tau_p\ll\tau_c \text{ and } |U_{mm'}(t)|\tau_p\ll 1.$$

A. Randomly spaced pulses

If the pulses occur randomly at some average rate γ, then they produce ''decoherence'' terms in the density-matrix equations of a form similar to those in [4],

$$\left(\frac{\partial\rho_{mm'}}{\partial t}\right)_{pulse}=-\gamma(1-\delta_{m,1}\delta_{m',-1} -\delta_{m,-1}\delta_{m',1}-\delta_{m,m'})\rho_{mm'}. \tag{7}$$

Here $\rho_{mm'}$ are the components of the active atom density matrix that have been averaged over the random phases produced by the external pulses.

Notice that only the coherences between the $m=0$ and $m=\pm 1$ experience any decay due to the pulses. In the laboratory frame, the polarization of the pulses $\hat{\boldsymbol{\epsilon}}$ defines the quantization axis for the active atom. Consequently, the pulse only couples the $|e,1,0\rangle$ state to the $|u,0,0\rangle$ state and only the $m=0$ substate amplitude acquires a random phase as a result of the pulse. One can also consider the situation in the collision frame in which the quantization axis is defined by $\hat{\mathbf{z}}'$ as shown in Fig. 2. In this coordinate system, the quantization axis is no longer parallel to the field polarization. However, Eqs. (7) still hold in the collision frame. When the field of the incident pulse has an arbitrary polarization relative to the atom and $\chi(t)/\delta(t)\ll 1$ where $\chi(t)=|E_0(t)|<e,J=1||d||u,J=0>/6\hbar$ is the reduced Rabi frequency, then the lowest-order effect of the pulse is simply a phase shift of each of the states. The $m=\pm 1$ states acquire the same phase shift, which is different than that of the $m=0$ state. Since the state vector of the active atom is only

defined up to an arbitrary global phase factor, one can factor out the random phase of the $m=\pm 1$ states. One is left with only the $m=0$ state experiencing a phase shift of $\Delta_s \tau_p$. There will be higher-order corrections proportional to $(\chi/\delta)^2$ that result in transitions between the $|e,1,m\rangle$ states. The density-matrix equations in the presence of randomly spaced pulses are

$$\frac{\partial \rho_{mm'}}{\partial t}=\sum_{m''}\left[iU_{m''m'}(t)\rho_{mm''}-iU_{mm''}(t)\rho_{m''m'}\right]+\left(\frac{\partial \rho_{mm'}}{\partial t}\right)_{pulse}. \tag{8}$$

B. Equally spaced pulses

Alternatively, one can consider the pulses to be equally spaced with the interval between the pulses denoted by $T=\gamma^{-1}$. The active atoms evolve freely according to the density matrix equations,

$$\frac{\partial \rho_{mm'}}{\partial t}=\sum_{m''}\left[iU_{m''m'}(t)\rho_{mm''}-iU_{mm''}(t)\rho_{m''m'}\right] \tag{9}$$

in the interval between pulses. When a pulse occurs, the coherence terms $\rho_{10}=\rho_{01}^*$ and $\rho_{-10}=\rho_{0-1}^*$ acquire a random phase, while the populations and $\rho_{-11}=\rho_{1-1}^*$ coherences are unaffected by the pulse. If one performs an ensemble average over the random phase, the boundary conditions after the nth pulse are

$$\rho_{10}(t_s+(n-1)T+\tau_p)=\rho_{-10}(t_s+(n-1)T+\tau_p)=0, \tag{10a}$$

$$\rho_{1-1}(t_s+(n-1)T+\tau_p)=\rho_{1-1}(t_s+(n-1)T), \tag{10b}$$

$$\rho_{mm}(t_s+(n-1)T+\tau_p)=\rho_{mm}(t_s+(n-1)T), \tag{10c}$$

where t_s is the time at which the first pulse occurs. The time at which the collision starts is arbitrary with respect to the pulse period T. Consequently, one must average over all times at which the first pulse could occur. The probability for the first pulse to occur in the interval (t_s,t_s+dt_s) is $\gamma\Theta(t_s)\Theta(T-t_s)dt_s$, where Θ is a unit step function.

In order to analyze the cross section for collisional transfer from the $m=0$ to $m=1$ sublevels in the presence of the external pulses, it is convenient to introduce the parameter

$$n_0=\gamma b_0/v=b_0/vT,$$

which represents the average number of pulses occurring in a collision having impact parameter equal to the Weisskopf radius.

III. CROSS SECTION FOR $n_0 \gg 1$

When $n_0 \gg 1$, there is a wide range of impact parameters for which the number of pulses per collision, $n(b)=n_0 b/b_0=\gamma\tau_c=\tau_c/T\gg 1$. The collisional evolution can be calculated using perturbation theory if $|U_{10}(t)\tau_c/n(b)|^2 n(b)\ll 1$. For randomly spaced pulses, in the perturbation theory limit, one can use Eqs. (7) and (8) to obtain the final-state population,

$$\rho_{11}(\infty)=\int_{-\infty}^{\infty}U_{10}(t')\int_{-\infty}^{t'}U_{01}(t'')e^{-\gamma(t'-t'')}dt'dt''+\text{c.c.}$$

$$=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}U_{10}(t')U_{01}(t'')e^{-\gamma|t'-t''|}dt'dt''. \tag{11}$$

For $\gamma\gg\tau_c^{-1}$, the exponential is very sharply peaked around $t'=t''$, $e^{-\gamma|t'-t''|}\approx\gamma^{-1}\delta(t'-t'')$ and

$$\rho_{11}(\infty)=\gamma^{-1}\int_{-\infty}^{\infty}|U_{10}(t)|^2dt, \tag{12}$$

a result that could have been written by inspection. Similarly, for equally spaced pulses, one can use Eqs. (9) and (10) to obtain

$$\rho_{11}(\infty)=T\int_{-\infty}^{\infty}|U_{10}(t)|^2dt. \tag{13}$$

One then obtains for the $m=1$ population after averaging over all collision geometries for $\gamma b/v=b/vT\gg 1$,

$$(\overline{\rho}_{11})_{pulses}=(\overline{\rho}_{11})_{no\ pulses}\frac{504}{84\pi}\frac{1}{\gamma(b/v)}$$

$$=(\overline{\rho}_{11})_{no\ pulses}\frac{504}{84\pi}\frac{1}{n_0(b/b_o)}, \tag{14}$$

where $(\overline{\rho}_{11})_{no\ pulses}$ is the perturbation theory result in the absence of external pulses. An explicit expression for $(\overline{\rho}_{11})_{no\ pulses}$ is given in Sec. IV.

Equation (14) can be used to estimate the cross section. We define the ratio of the $m=0\rightarrow m=1$ cross sections with and without pulses as $S(n_0)$,

$$S(n_0)=\frac{(\sigma)_{pulses}}{(\sigma)_{no\ pulses}}=\int_0^{\infty}(\overline{\rho}_{11})_{pulses}(y,n_0)y\,dy\Big/\int_0^{\infty}(\overline{\rho}_{11})_{no\ pulses}(y)y\,dy, \tag{15}$$

where $y=b/b_o$. To estimate $S(n_0)$ for $n_0\gg 1$ we use the perturbation theory result for $y>y'$, where y' is defined by $\overline{\rho}_{11}(y')=1$, and the strong collision, asymptotic average value, $\overline{\rho}_{11}=4/15$ [12] for $0<y<y'$. In this manner, we obtain

$$S(n_0)=\frac{28}{31}\left(\frac{1.91}{n_0}\right)^{2/11}. \tag{16}$$

The power law $S(n_0)\sim n_0^{-2/11}$ is a general result that holds for all potentials that vary as $1/R^6$ and was previously de-

rived in [4]. This result clearly indicates that as $n_0 \to \infty$ the atom is frozen in its initial state.

IV. CROSS SECTION FOR $n_0 \sim 1$

When $n_0 \sim 1$, it is necessary to solve the appropriate equations numerically for *all* collision orientations and then average the results. Since this procedure is rather time consuming, we adopt an alternative method that should give results that agree at least qualitatively with the exact ones. A ''typical collision'' geometry is defined in which, in the absence of external pulses *and in the perturbation theory limit*, the value of ρ_{11} calculated for the typical collision geometry equals the value of $\overline{\rho_{11}}$.

In perturbation theory, with $a_0(-\infty)=1$, one finds from Eqs. (2) and (6) that

$$\begin{aligned}\rho_{11}(\alpha,\beta,\xi)_{no\ pulses} &= |a_1(\infty)|^2 \\ &= |A|^2 \left| \sum_m \int_{-\infty}^{\infty} D^{(2)}_{-1m}(\alpha,\beta,\xi) \frac{Y^*_{2,m}(\theta(t),\phi(t))}{R(t)^6} dt \right|^2 \\ &= \frac{5\pi|A|^2}{16^3 b^{10} v^2} \left\{ \frac{3}{4}(1-\cos 4\alpha) + \frac{3}{4}(163+\cos 4\alpha \right. \\ &\quad - 36\cos 2\xi \cos 2\alpha)\cos^2\beta + 27\sin\beta\cos\beta \\ &\quad \left. \times \sin 2\xi \sin 2\alpha \right\} \sin^2\beta, \end{aligned} \tag{17}$$

where

$$|A|^2 = \frac{1}{15\pi}\left(\frac{5|C|^2}{4}\right)^2.$$

Using the orthogonality relationship for the rotation matrices $D^{(j)}_{m'm}(\alpha,\beta,\xi)$ with respect to integration over rotation angles α,β,ξ, one obtains the orientation averaged population

$$(\overline{\rho_{11}})_{no\ pulses} = \frac{21\pi|A|^2}{1024 b^{10} v^2}. \tag{18}$$

By introducing the quantity $F(\alpha,\beta,\xi)$ defined as

$$\rho_{11}(\alpha,\beta,\xi)_{no\ pulses} = (\overline{\rho_{11}})_{no\ pulses} F(\alpha,\beta,\xi), \tag{19}$$

the ''typical collision'' can be defined as one for which $F(\alpha,\beta,\xi)=1$. In this manner, the transition probability produced by a perturber with orientation (α,β,ξ) relative to our standard collision geometry in the perturbation theory limit is the same as that produced *on average* by a collision. For $\alpha=\xi=0$ and $\beta=\pi/6.25$ one finds that $\rho_{11}(0,\pi/6.25,0) = 1.018\overline{\rho_{11}}$.

The choice $\alpha=\xi=0$ has the added benefit of converting the three-level problem into an effective two-level problem. The collision matrix for the ''typical collision'' geometry $U_{mm'}(t,0,\beta,0)$ is real for all m and m'. If one makes a change of basis to a symmetric-antisymmetric basis given by

$$\begin{aligned} a_0 &= a_0, \\ a_+ &= (a_{+1} + a_{-1})/\sqrt{2}, \\ a_- &= (a_{+1} - a_{-1})/\sqrt{2}, \end{aligned} \tag{20}$$

the Schrödinger equation in the new basis is

$$i\dot{a}_+ = \{U_{11}(t,0,\beta,0) + U_{1-1}(t,0,\beta,0)\}a_+, \tag{21a}$$

$$\begin{aligned} i\dot{a}_- &= \{U_{11}(t,0,\beta,0) - U_{1-1}(t,0,\beta,0)\}a_- \\ &\quad + \sqrt{2}U_{10}(t,0,\beta,0)a_0, \end{aligned} \tag{21b}$$

$$i\dot{a}_0 = \sqrt{2}U_{10}(t,0,\beta,0)a_- + U_{00}(t,0,\beta,0)a_0. \tag{21c}$$

Here explicit use has been made of the relationship $U_{-m-m'} = (-1)^{m-m'} U^*_{mm'}$. The symmetric state a_+ decouples from the other two states and simply acquires an overall phase. Moreover, for the initial conditions $a_0(-\infty)=1$ and $a_{-1}(-\infty)=a_{+1}(-\infty)=0$, $\rho_{11}(\infty)$ is completely independent of $a_+(\infty)$. It is now a simple matter to incorporate the external pulses into these equations.

A. Randomly spaced pulses

For randomly space pulses, Eqs. (7), (8), (20), and (21) can be recast in the form of Bloch equations,

$$\dot{w} = 2\sqrt{2}U_{10}(t,0,\beta,0)v, \tag{22a}$$

$$\begin{aligned} \dot{v} &= (U_{11}(t,0,\beta,0) - U_{1-1}(t,0,\beta,0) - U_{00}(t,0,\beta,0))u \\ &\quad - 2\sqrt{2}U_{10}(t,0,\beta,0)w - \gamma v, \end{aligned} \tag{22b}$$

$$\dot{u} = -(U_{11}(t,0,\beta,0) - U_{1-1}(t,0,\beta,0) - U_{00}(t,0,\beta,0))v - \gamma u, \tag{22c}$$

with

$$\begin{aligned} w(t) &= \rho_{--}(t) - \rho_{00}(t), \\ v(t) &= i(\rho_{-0}(t) - \rho_{0-}(t)), \\ u(t) &= \rho_{-0}(t) + \rho_{0-}(t). \end{aligned} \tag{23}$$

In terms of a dimensionless time $x = tv/b$ and a coupling constant $\eta = 25|C|^2/(40\pi b^5 v) = (b_0/b)^5$ one has

$$dw/dx = \eta f(x,\beta)v, \tag{24a}$$

$$dv/dx = \eta g(x,\beta)u - \eta f(x,\beta)w - n_0\eta^{-1/5}v, \tag{24b}$$

$$du/dx = -\eta g(x,\beta)v - n_0\eta^{-1/5}u, \tag{24c}$$

$$f(x,\beta) = \frac{2x\cos 2\beta - (1-x^2)\sin 2\beta}{(1+x^2)^4}, \tag{24d}$$

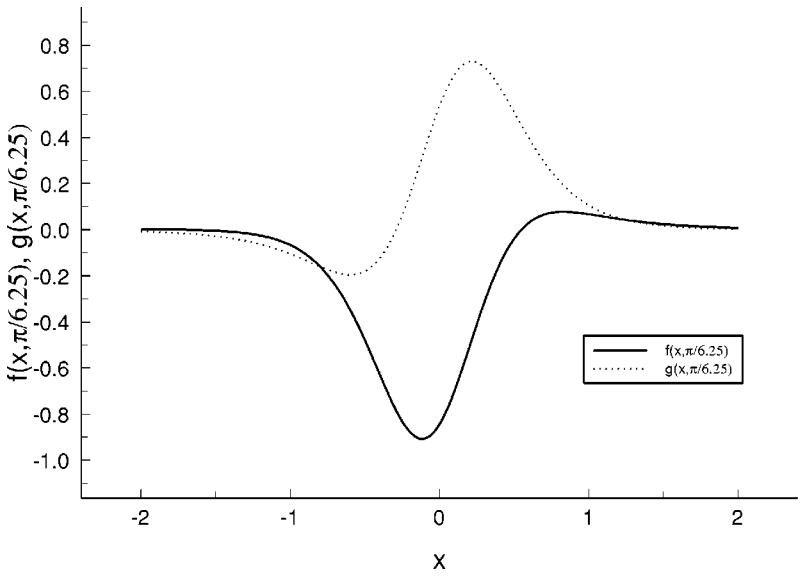

FIG. 3. Plot of $f(x,\pi/6.25)$ and $g(x,\pi/6.25)$ as a function of x.

$$g(x,\beta)=\frac{2x\sin 2\beta+(1-x^2)\cos 2\beta}{(1+x^2)^4}, \tag{24e}$$

$$w(-\infty)=-1;v(-\infty)=u(-\infty)=0. \tag{24f}$$

Figure 3 shows a plot of $f(x,\pi/6.25)$ and $g(x,\pi/6.25)$. For fixed η and β one can think of ηf and ηg as effective Rabi frequencies and detunings for the system.

In general, to find $\rho_{11}(t)$, one needs to know $w(t)$ as well as $\rho_{+-}(t)$ and $\rho_{++}(t)$, since

$$\rho_{11}(t)=\left(\frac{a_+ + a_-}{\sqrt{2}}\right)\left(\frac{a_+ + a_-}{\sqrt{2}}\right)^* =\frac{w(t)+1}{4}+\frac{\rho_{++}(t)+\rho_{+-}(t)+\rho_{-+}(t)}{2}. \tag{25}$$

However, for the initial conditions $a_0(-\infty)=1$ and $a_{+1}(-\infty)=a_{-1}(-\infty)=0$ it is easy to show that $\rho_{++}(t)=\rho_{+-}(t)=0$ for all t. Therefore, as asserted earlier, $\rho_{11}(t)$ is independent of $a_+(t)$ and is given by $\rho_{11}(t)=[w(t)+1]/4$.

Equations (24) can be integrated numerically with respect to x for fixed η and β. For each η the Bloch equations were integrated over the interval $-3\leqslant x\leqslant 3$, which is taken to simulate the interval $(-\infty,\infty)$. The numerical integration over η (or b), needed to evaluate $S(n_0)$, was done for $0.01\leqslant\eta\leqslant 200$ with $\beta=\pi/6.25$. In Fig. 4 a plot of $\rho_{11}(x=3)$ for $n_0=2$ is shown as a function of b/b_0. Results from a numerical evaluation of $S(n_0)$ [Eq. (15)] are shown in Fig. 5. One sees that for $n_0=1$, one achieves a 15% reduction in the cross section and, for $n_0=4$, the reduction is 36%. This shows that even a moderate number of pulses can result in dramatic reductions in the cross section.

In the case of $n_0=0$, one can compare the cross section obtained for the ''typical collision'' geometry with the cross

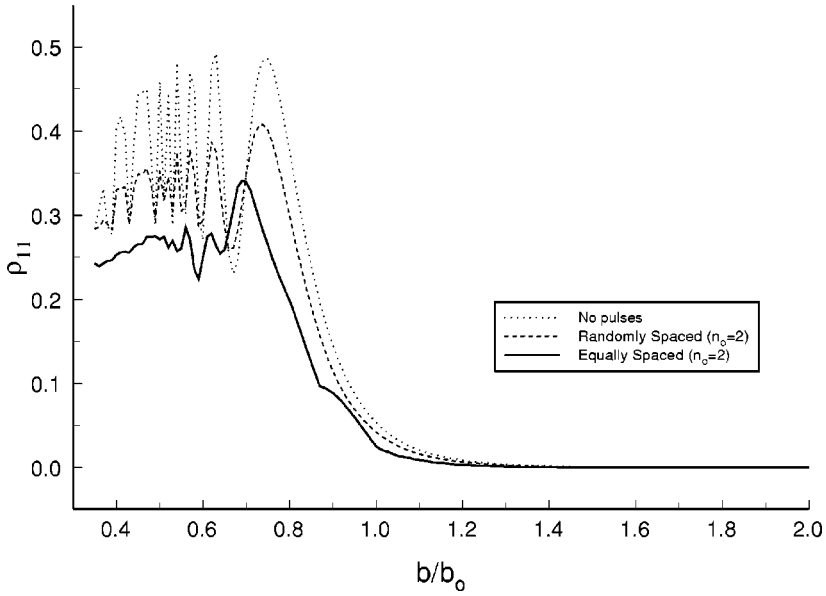

FIG. 4. Plot of $\rho_{11}(x=3)$ as a function of $b/b_0=\eta^{-1/5}$ for three cases: (i) no pulses; (ii) $n_0=2$ for randomly spaced pulses; and (iii) $n_0=2$ for equally spaced pulses.

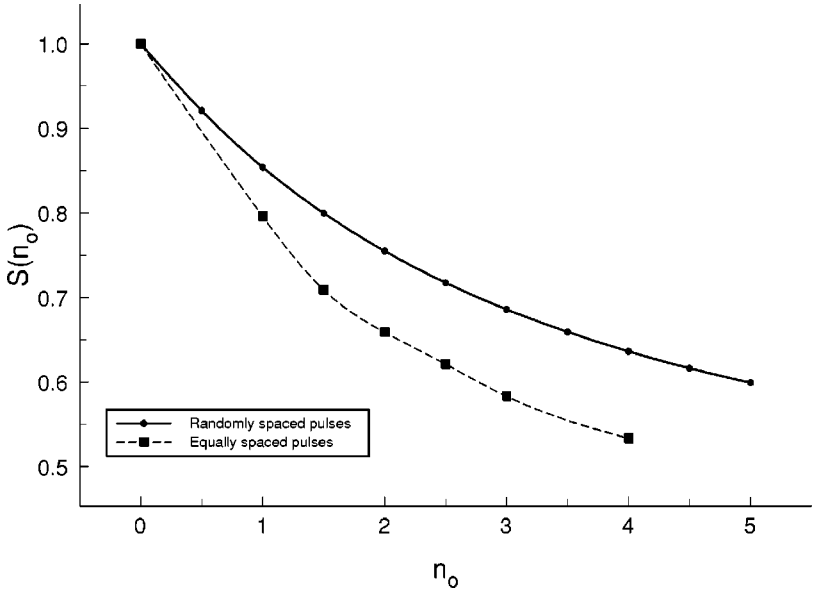

FIG. 5. Plot of $S(n_0)$ for randomly spaced pulses and equally spaced pulses.

section (averaged over all collision geometries) calculated in [12]. One finds that there is a difference of only 0.1% [13], which is in fact less than the numerical error quoted in [12] and the numerical error of our results. This level of agreement must be regarded as fortuitous. Regardless, it shows that the ''typical collision'' geometry is a viable alternative to a complete averaging for the $m=0\rightarrow m=1$ transition. It is important to note that the typical collision geometry was defined by matching *perturbation theory* results—there was no guarantee it would yield quantitatively accurate results when the average over *all* impact parameters was carried out.

B. Equally spaced pulses

Now consider what happens when the pulses occur at well-defined times with each pulse occurring at intervals $T=\gamma^{-1}$, with $n_0=\gamma b_0/v$. To solve this problem one sets $n_0=0$ in Eqs. (24) so that the Bloch equations are

$$dw/dx=\eta f(x,\beta)v, \tag{26a}$$

$$dv/dx=\eta g(x,\beta)u-\eta f(x,\beta)w, \tag{26b}$$

$$du/dx=-\eta g(x,\beta)v. \tag{26c}$$

The Bloch vector evolves freely from its initial condition until x_s, which is the time of the first pulse. Since the pulse completely randomizes the coherences, the pulse projects the Bloch vector back on the w axis. Thus one has the new initial conditions $w(x_s^+)=w(x_s^-)$ and $v(x_s^+)=u(x_s^+)=0$. The Bloch vector again evolves freely until the time $x_s+Tv/b=x_s+n_0^{-1}\eta^{1/5}$ when the next pulse occurs. This pulse again projects the vector back on the w axis and the procedure repeats until the end of the collision. It is also necessary to average over the time of the first pulse that has a probability density of $n_0\eta^{-1/5}\Theta(x_s)\Theta(n_0^{-1}\eta^{1/5}-x_s)$. The results are also plotted in Fig. 5. For $n_0=1$, one achieves a 20% reduction in the cross section and when $n_0=4$ the reduction is 47%. Notice that $S(n_0)$ does not exhibit any oscillations, as in [4]. The oscillations were a consequence of the collision area between any two pulses always being the same for a collision interaction modeled as a square pulse. This does not occur for our smooth potential.

It is interesting to consider how x_s affects $\rho_{11}(\infty)$. When $n_0(b/b_0)\gg 1$, the affect of the first pulse is negligible since it occurs in the far wing of the collision. However, when only a single pulse occurs during the collision, one can clearly see the effect that the pulse has in disrupting the coherent evolution of the active atom due to the collision. We consider the case of $b=b_0$ and plot $\rho_{11}(\infty)$ as a function of x_s in Fig. 6. The population $\rho_{11}(\infty)$ is a minimum when x_s corresponds to the time when $|f(x,\pi/6.25)|$ is largest. Since $\eta f(x,\beta)$ plays the role of a Rabi frequency, we see that the pulse effectively destroys the coherent evolution of the system from $m=0$ to $m=1$. Similarly, $\rho_{11}(\infty)$ achieves its maximum value when x_s coincides with the maximum of $g(x,\pi/6.25)$. Recall that $\eta g(x,\beta)$ is the detuning for our two-level system. The pulse effectively reduces the detuning and thereby enhances the transition amplitude.

It should be emphasized that this typical collision works best for the particular scattering process $m=0\rightarrow m=1$ and would yield poorer results if used to try to determine other elements in the scattering superoperator for the active atom density matrix. For example, to compute the cross section for scattering from $m=-1\rightarrow m=+1$, one cannot ignore the $\rho_{++}(t)$ and $\rho_{+-}(t)$ terms. Finally, note that the standard geometry ($\alpha=\gamma=\xi=0$) cannot be used as our typical collision geometry since the cross section for this geometry, in the absence of pulses is only 60% of the averaged cross section [12]. The cross section is particularly small owing to the fact that $g(x,0)$ is even while $f(x,0)$ is odd with respect to time reversal.

V. DISCUSSION

The application of the external pulses shows a suppression of the transition probabilities between different mag-

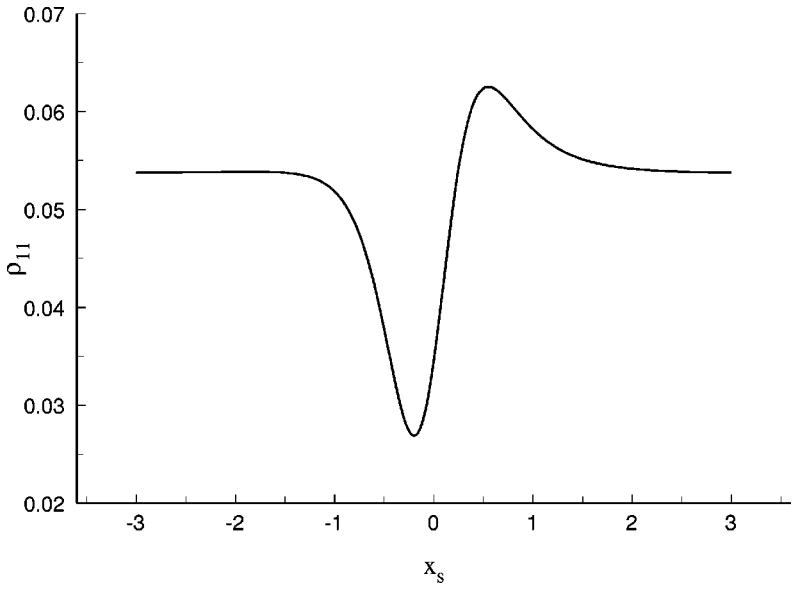

FIG. 6. A plot of $\rho_{11}(x=3,b=b_0)$ when a single pulse occurs at $x=x_s$ during the collision.

netic substates. The magnetic substates are defined with respect to a quantization axis that is arbitrary in the absence of an external field. For any particular collision in the presence of pulses, there are two convenient ways to define the quantization axis. One could define the quantization axis as the $\hat{\mathbf{z}}'$ axis in the collision frame or by the polarization $\hat{\boldsymbol{\epsilon}}$ of the linearly polarized external pulses in the laboratory frame. Let us consider the former case. When one rotates to a different collision orientation, one must also rotate the field polarization $\hat{\boldsymbol{\epsilon}}$ and $\mathbf{k}$ vector in the appropriate way. Symbolically, if $\rho_0(\mathbf{E}_p)_{mm'}$ represents the density-matrix solution in the collision frame with an external field $\mathbf{E}_p$ representing the pulses, then the solution for an arbitrary collision geometry represented by Ω is

$$\rho(\mathbf{E}_p,\Omega)_{mm'}=D^{(J)}_{mn}(\Omega)^{-1}\rho_0(R_a(\Omega)^{-1}\mathbf{E}_p)_{nq}D^{(J)}_{qm'}(\Omega). \tag{27}$$

Here $D^{(J)}_{mn}(\Omega)$ is an active rotation matrix element and $R_a(\Omega)$ is the representation of the corresponding active rotation acting on $\mathbf{E}_p$. Physically, this corresponds to a system in which the perturber's trajectory relative to the active atom is related to the trajectory in the collision frame, $\mathbf{R}(t)=vt\hat{\mathbf{x}}'+b\hat{\mathbf{z}}'$, by the rotation $R_a(\Omega)$ while the orientation of the external pulses relative to the active atom are unchanged.

Since there is a functional dependence on Ω in $\rho_0(R_a(\Omega)^{-1}\mathbf{E}_p)$, averaging over all Ω is nontrivial since one cannot take advantage of the properties of $D^{(J)}_{mn}(\Omega)$ to obtain a simple form for the averaged superscattering matrix as in [12]. Consequently, we are led to the idea of a ''typical collision'' geometry in order to avoid these problems and still obtain useful numerical results. A similar approach is employed in [5] when calculating the scattering matrix. They calculate their cross sections in the laboratory frame (with quantization axis $\hat{\boldsymbol{\epsilon}}$) as a function of the angles that characterize the collision orientation in the laboratory frame and use a single polar angle that preserves the collision characteristics instead of numerically averaging over the full range of angles.

In our analysis, the pulse that initially excited the population in $|e,1,0\rangle$ was linearly polarized in the $\hat{\mathbf{z}}$ direction of the laboratory frame. If, instead the excitation pulse has arbitrary polarization, then it will create a coherent superposition $|\psi(0)\rangle=\Sigma_m a_m(0)|e,1,m\rangle$. If this state can be transformed into an $m'=0$ state with respect to the quantization axis $\hat{\mathbf{z}}'$, in a rotated laboratory system, then the above analysis remains valid in the new coordinate system.

Thus one can preserve coherent superpositions of states, provided that there exists a coordinate system $(\hat{\mathbf{x}}',\hat{\mathbf{y}}',\hat{\mathbf{z}}')$ related to the original laboratory frame $(\hat{\mathbf{x}},\hat{\mathbf{y}},\hat{\mathbf{z}})$ by a rotation parameterized by Ω', such that

$$|e,1,0'\rangle=U_p(\Omega')|\psi(0)\rangle, \tag{28a}$$

$$|\psi(0)\rangle=U_a(\Omega')|e,1,0'\rangle=\sum_m D^{(1)}_{m0}(\Omega')|e,1,m\rangle. \tag{28b}$$

Here $|e,1,0'\rangle$ is the $m'=0$ state with respect to the $(\hat{\mathbf{x}}',\hat{\mathbf{y}}',\hat{\mathbf{z}}')$ axes and $U_p(\Omega')$ represents the *passive* rotation acting on our state while $U_a(\Omega')=U_p^{-1}(\Omega')$ is the corresponding active rotation of the system [14]. As an example, x-polarized light with $\mathbf{k}=k\hat{\mathbf{z}}$ will excite the state $|\psi(0)\rangle=(1/\sqrt{2})(|e,1,-1\rangle-|e,1,+1\rangle)=|e,1,0'\rangle$ for $\hat{\mathbf{z}}'=\hat{\mathbf{x}}$. The external pulses prevent decoherence of this superposition state.

The important effect being demonstrated here is that very rapid perturbations of a system that destroy the phase relationship between the amplitudes $a_m(t)$ at t and $t+\delta t$, inhibit the coherent time evolution of the states. The idea bears a close resemblance to the quantum Zeno effect [15] in which the coherent evolution of a quantum system is inhibited by invoking the measurement postulate to project the state vec-

tor onto an eigenstate of the measurement operator. Although the net result is the same, the process described in this paper does not qualify as an example of the quantum Zeno effect [4]. Instead our proposal is similar in spirit to the classical Drude theory of conduction in metals, where the electrons undergo randomizing collisions with ions that cause them to lose all information about their velocity before the collision [16].

The effect being proposed in this paper can be understood in the more general context of system plus reservoir interactions. One can consider the foreign gas perturbers to represent a thermal bath having correlation time τ_c. Similarly, one can attribute the random phase factors produced by the pulses as due to the coupling of the active atom to a second reservoir with a correlation time $\tau_{c'}$ such that $\tau_{c'} \ll \tau_c$. The interaction of the active atom S with the two reservoirs $B1$ and $B2$ can be described formally by the total Hamiltonian H and state vector $|\psi(t)\rangle$,

$$H = H_S + H_{B1} + H_{B2} + H_{SB1} + H_{SB2}, \tag{29}$$

where H_S is the active atom Hamiltonian and H_{B1} is the reservoir of foreign gas perturbers. The Hamiltonian $H_{SB1} = g_1 \Sigma_\alpha S_\alpha \otimes B_\alpha^{(1)}$ is the interaction between the active atom and the bath of perturbers, and S_α are the operators acting on the active atom Hilbert space and have nonzero off-diagonal elements. The Hamiltonian H_{B2} represents the second reservoir with correlation time $\tau_{c'}$, and $H_{SB2} = g_2(t) \Sigma_\alpha S'_\alpha \otimes B_\alpha^{(2)}$ is the interaction of the active atom with the reservoir due to the pulses. The operators S'_α that act on the Hilbert space of the active atom are diagonal in the representation considered. If the coupling of S to $B2$ is of the form $g_2(t) = G \Sigma_n \Theta(t - nT) \Theta(nT + \tau_p - t)$ and $\tau_{c'} < \tau_p \ll T < \tau_c$ where G is arbitrarily large so that the terms H_S and H_{SB1} can be neglected when $g_2(t) \neq 0$, then the effect of the coupling to the bath at $t = nT + \tau_p$ is

$$|\psi(nT + \tau_p)\rangle = \exp\left[-\frac{i}{\hbar} \left(G \tau_p \sum_\alpha S'_\alpha \otimes B_\alpha^{(2)} + \tau_p H_{B1} + \tau_p H_{B2} \right) \right] |\psi(nT)\rangle. \tag{30}$$

The basis states for the Hilbert space of the active atom simply acquire a phase due to the interaction with $B2$. Moreover, since $\tau_p > \tau_{c'}$ the phase of these states at $nT + \tau_p$ will be uncorrelated with the phase at nT. The random phases from the optical pulses can be thought of as the periodic coupling to a very fast bath. Thus our proposal involves the use of one reservoir to inhibit transitions induced by another reservoir which has a much longer correlation time.

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