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Elliptical billiard-ball echo model

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The billiard-ball echo model is expanded to include the effects of using sub-Doppler-linewidth lasers in photon-echo experiments. The simple heuristic model which emerges explains such echo phenomena in gases as the echo time width and the degradation of echo intensity with angling of the excitation pulses. We derive simple results in the small-pulse-area limit and give formal expressions for excitation pulses of arbitrary area.

INTRODUCTION

In a recent paper[1] an unorthodox method was presented for analyzing optical coherent transient phenomena in gaseous media. Called the billiard-ball echo model it began by localizing the gaseous atoms with atomic wave packets. When these wave packets were irradiated by a series of light pulses they split into separate parts corresponding to each of the atomic states generated. Each resulting wave packet recoiled according to whether it absorbed or emitted a photon while being formed. Whenever overlap occurred between two daughter wave packets of the same parent, coherent radiation would ensue and the various photon-echo phenomena were readily explained and visualized.

This billard-ball echo model was developed to obtain a simple analytical tool which would work equally well for the two-pulse photon echo and multiple-pulse—multiple-level echoes. It was specialized to short-pulse excitations which uniformly excited all atoms independent of their velocity thus giving rise to spherical wave packets which were called billiard balls. Long-pulse excitation leads to a more complicated shape which we analyze herein. As demonstrated in the original paper for the spherical wave packet and similarly here for the more general case, wave-packet spreading can be neglected. In the body of this paper we therefore deal only with the unspread wave packets, which makes the presentation much cleaner and easier to follow. Long-pulse excitations do not change the essential features of the billiard-ball echo theory. Even though the shape of the excited-state wave packets is no longer spherical, the recoil or billiard-ball analysis remains the same. Only questions of radiated field amplitude need be addressed as they depend on the shape of the colliding wave packets.

The short-pulse analysis has certain features which we can carry over to the more general case. It is convenient to think of the excitation pulse as arriving at an instant in time, with the wave function and wave packet being instantly transformed. We must, of course, require that we obtain the correct final state and we may not use these wave functions while the excitation pulse is being applied. As we are primarily interested in echo phenomena this restriction is of no consequence.

WAVE-PACKET GENERATION

We irradiate a gaseous sample with an electric field pulse of area Θ, wave vector $\vec{k}$, central frequency ω, and envelope $E(t-\vec{k}\cdot\vec{r}/ck)$. Let ω be at exact resonance with an atom which is initially stationary. If $\hbar\Omega$ is the energy separation of the atom's ground ($|1\rangle$) and excited ($|2\rangle$) states ω must be

$$\omega=\Omega+\frac{\hbar k^2}{2m}\ , \tag{1}$$

where m is the atomic mass and the second term is the recoil energy.

The envelope E has a duration which is nonzero but finite. The pulse therefore does not excite all atoms uniformly but rather to an extent depending on each atom's Doppler shift from exact resonance. For an atom with momentum $\hbar\vec{q}$ this shift is $\hbar\vec{k}\cdot\vec{q}/m$.

In analyzing the effect of an excitation pulse on our gas we work in a scheme which replaces the actual pulse with an equivalent instantaneous pulse. This short pulse is defined by its effect on the atomic wave functions which after passage of the actual excitation pulse must be identical to what the actual excitation pulse would have produced. Taking this pulse to arrive at $\vec{r}=0$ at time $t=0$ we describe its effect on an "atom" initially localized at the origin by writing the wave packets just before and after the passage of the pulse,

$$|\Psi(t=0^-)\rangle = \int d\vec{q}\, A(\vec{q})\,|\vec{q}\rangle\,|1\rangle \tag{2}$$

and

$$|\Psi(t=0^+)\rangle = \int d\vec{q}\, A(\vec{q})[\cos(\theta/2)\,|\vec{q}\rangle\,|1\rangle + i\sin(\theta/2)e^{i\phi}\,|\vec{q}+\vec{k}\rangle\,|2\rangle] \,. \tag{3}$$

$|\vec{q}\rangle$ is a center-of-mass momentum eigenstate. In this instantaneous treatment θ and ϕ assume their final values immediately instead of evolving during the time the pulse is actually present. The wave packets are constructed from momentum states with a Maxwellian distribution:

$$A^2(\vec{q}) = \frac{1}{(\pi q_0^2)^{3/2}} e^{-\vec{q}^{\,2}/q_0^2} , \tag{4}$$

where

$$q_0 = \left[\frac{2mk_b T}{\hbar^2} \right]^{1/2} . \tag{5}$$

The parameters θ and ϕ specify the excited state associated with the momentum component $\hbar\vec{q}$. As this can depend only on the associated Doppler shift we write

$$\theta = \theta(q_k) \,, \quad \phi = \phi(q_k) \,, \tag{6}$$

where q_k is the component of $\vec{q}$ along $\vec{k}$,

$$\vec{q} = q_\perp \hat{\perp} + q_k \hat{k} \tag{7}$$

and $\hat{\perp}$ is a generalized unit vector perpendicular to $\hat{k}$. For $q_k = 0$, $\theta = \Theta$ and $\phi = 0$.

Reexpressing $|\Psi(t=0^+)\rangle$ in terms of effective distribution functions $A_1(\vec{q})$ and $A_2(\vec{q})$, we write

$$|\Psi(t=0^+)\rangle = \cos\frac{\Theta}{2} \int d\vec{q}\, A_1(\vec{q})\,|\vec{q}\rangle\,|1\rangle + i\sin\frac{\Theta}{2} \int d\vec{q}\, A_2(\vec{q})\,|\vec{q}+\vec{k}\rangle\,|2\rangle \,, \tag{8}$$

where

$$A_1(\vec{q}) = \frac{\cos[\theta(q_k)/2]}{\cos(\Theta/2)} A(\vec{q}) \tag{9}$$

and

$$A_2(\vec{q}) = \frac{\sin[\theta(q_k)/2]}{\sin(\Theta/2)} e^{i\phi(q_k)} A(\vec{q}) \,. \tag{10}$$

Since $\vec{q}^{\,2} = q_\perp^2 + q_k^2$ the A's factor further and we can write

$$A_{1,2}(\vec{q}) = \frac{1}{(\pi q_0^2)^{1/2}} e^{-q_\perp^2/2q_0^2} a_{1,2}(q_k) \,, \tag{11}$$

where

$$a_1(q_k) = \frac{1}{(\pi q_0^2)^{1/4}} e^{-q_k^2/2q_0^2} \frac{\cos[\theta(q_k)/2]}{\cos(\Theta/2)} \tag{12}$$

and

$$a_2(q_k) = \frac{1}{(\pi q_0^2)^{1/4}} e^{-q_k^2/2q_0^2} \times \frac{\sin[\theta(q_k)/2]}{\sin(\Theta/2)} e^{i\phi(q_k)} \,. \tag{13}$$

The coherently radiated field with amplitude $\vec{E}_R$ at frequency ω and wave vector $\vec{k}_R$ generated by a dipole distribution $\vec{P}$ oscillating at ω is calculated from[2]

$$\vec{E}_R = \hat{e} k_R^2 \frac{e^{-ik_R R}}{R} \int d\vec{r}\, P e^{i\vec{k}_R\cdot\vec{r}} \,, \tag{14}$$

where we have assumed the spatial extent of the source is small compared to R, the distance from the source to where the field is measured. $P\hat{e}$ is defined by $(\vec{R}\times\vec{P})\times\vec{R} = \hat{e}PR^2$, $\hat{e}$ being a unit vector. If the volume of the source is large and the dipole distribution is characterized by a k vector $\vec{k}$, then $\vec{E}_R$ is dominated by $\vec{k}_R = \vec{k}$. We accordingly restrict ourselves to this condition just as in Ref. 1. The radiative moment $Pe^{i\vec{k}\cdot\vec{r}}$ is to be calculated throughout the sample volume but as shown in Ref. 1 it suffices to consider a single wave packet initially centered at $\vec{r}=0$ in performing this calculation as all wave packets contribute in identical manner. In order to calculate the integrated radiative moment we must construct spatial wave packets by forming

$$\langle \vec{r}\,|\,\vec{q}\rangle = \frac{1}{(2\pi)^{3/2}} e^{i\vec{q}\cdot\vec{r}} \tag{15}$$

and then follow these wave packets in time. With time the wave packets spread out and the excited state recoils with velocity $\hbar\vec{k}/m$. Just as in Ref. 1 we can neglect spreading since it has no effect on the radiative moment. We can then write the spatial wave packet at time t in the form

$$\Psi(t) = \cos\frac{\Theta}{2} F_1(\vec{r})\,|1\rangle + i\sin(\Theta/2) e^{-i(\omega t - \vec{k}\cdot\vec{r})} F_2\left[\vec{r} - \frac{\hbar\vec{k}}{m} t\right] |2\rangle \,, \tag{16}$$

where

$$F_{1,2}(\vec{r}) = \frac{1}{(2\pi)^{3/2}} \int d\vec{q}\, A_{1,2}(\vec{q}) e^{i\vec{q}\cdot\vec{r}} \,. \tag{17}$$

Using Eq. (14) and retaining only those terms that contribute to the macroscopically radiated field we finally have

$$\vec{E}_R = \hat{e}\frac{k^2}{R}e^{-i\vec{k}\cdot\vec{R}}N\cos\left[\frac{\Theta}{2}\right]\sin\left[\frac{\Theta}{2}\right]P_{21} \times \int d\vec{r}F_1(\vec{r})F_2^*\left[\vec{r}-\frac{\hbar\vec{k}}{m}t\right], \quad (18)$$

where we have taken $\vec{R}$ along $\hat{k}$ and have written P_{21} for the expectation value $\langle 2|\hat{P}|1\rangle$ of the dipole moment operator. The number of atoms N appears as a factor since each wave packet contributes in identical fashion. This form of $\vec{E}_R$ is identical to that obtained in Ref. 1.

Since F_1 and F_2 are the wave-packet amplitudes we find the radiated field to be proportional to the overlap of the ground- and excited-state wave packets. These wave packets separate at the recoil velocity $\hbar\vec{k}/m$, which is a function of the atomic mass and transition frequency only. The shape of the wave packets therefore determines the time evolution of $\vec{E}_R$. $\vec{E}_R$ can be calculated from Eq. (18) only after the excitation has passed but for calculating photon-echo amplitudes this poses no problem. For free decay calculations we are restricted to short-pulse excitations.

WAVE-PACKET SHAPE (SHORT PULSE)

Before proceeding to calculate the echo signal let us examine the shape of the atomic wave packets. This is important in as much as the echo signal amplitude and duration are sensitive to wave-packet shape. First we consider the short-pulse limit where the action of an excitation pulse is independent of atomic velocity. In this case $\theta(q_k)$ and $\phi(q_k)$ are independent of q_k with

$$\theta(q_k)=\Theta \quad (19)$$

and

$$\phi(q_k)=0\ . \quad (20)$$

The effective distribution functions $A_1(\vec{q})$ and $A_2(\vec{q})$ are then both given by $A(\vec{q})$ and so the wave-packet amplitudes $F_1(\vec{r})$ and $F_2(\vec{r})$ are equal. Using Eqs. (17) and (4) we obtain

$$F_1(\vec{r})=F_2(\vec{r}) = \frac{1}{(2\pi)^{3/2}}\frac{1}{(\pi q_0^2)^{3/4}}\int d\vec{q}\,e^{-\vec{q}^{\,2}/2q_0^2}e^{i\vec{q}\cdot\vec{r}} = \left[\frac{q_0}{\sqrt{\pi}}\right]^{3/2}e^{-q_0^2\vec{r}^{\,2}/2}\ . \quad (21)$$

This is the result of Ref. 1: The wave packets are spherical with a Gaussian amplitude falloff from their centers. A measure of their size is the radius r at which the wave-packet intensity is down by a factor of $1/e$,

$$r_{1/e}=\frac{1}{q_0}\ . \quad (22)$$

This is just the de Broglie or thermal wavelength. A measure of the time τ_D for which the ground- and excited-state wave packets maintain appreciable overlap is obtained by setting this wave-packet radius q_0^{-1} equal to the magnitude of the recoil velocity $\hbar k/m$ multiplied by the separation time τ_D. Thus

$$\frac{\hbar k}{m}\tau_D=\frac{1}{q_0}\ . \quad (23)$$

But this may be rearranged to read

$$\frac{\hbar q_0}{m}\tau_D=\frac{1}{k} \quad (24)$$

which is the requirement that the Doppler velocity times τ_D must be an optical wavelength. Thus the billiard-ball echo model leads directly to what would have been obtained by using the standard Doppler dephasing arguments.[3]

WAVE-PACKET SHAPE (LONG PULSE)

We now consider long-pulse excitation where $A_1(\vec{q})$ and $A_2(\vec{q})$ are not identical and differ from $A(\vec{q})$. However, as shown in Eq. (11), they continue to share the feature of an identical dependence on $q_\perp$. This carries over to the wave-packet amplitudes which are the Fourier transforms of $A_1(\vec{q})$ and $A_2(\vec{q})$. It follows that

$$F_{1,2}(\vec{r})=\left[\frac{q_0}{\sqrt{\pi}}\right]e^{-q_0^2 r_\perp^2/2}f_{1,2}(r_k)\ , \quad (25)$$

where

$$f_{1,2}(r_k)=\frac{1}{(2\pi)^{1/2}}\int dq_k a_{1,2}(q_k)e^{iq_k r_k}\ . \quad (26)$$

As with $q_\perp$ and q_k, $r_\perp$ and r_k are the components of $\vec{r}$ along $\hat{\perp}$ and $\hat{k}$.

The transverse character of the wave packets is therefore the same, with an extent of order q_0^{-1}. However, the wave packets become elongated along $\hat{k}$.

WAVE-PACKET SHAPE (LONG PULSE, SMALL AREA): ELLIPITICAL BILLARD BALLS

That the wave packets become elongated along k may be seen by considering the small area limit where each atom is only excited in proportion to

$$\theta(q_k)=\frac{1}{\hbar}P\int dt\,E(t)\cos[(\omega-\omega_{q_k})t]\ , \tag{27}$$

the Fourier component of the radiation field at the atom's Doppler-shifted resonance frequency,

$$w_{q_k}=\omega+\hbar\vec{k}\cdot\vec{q}/m\ . \tag{28}$$

In the small-angle limit the angle $\phi(q_k)$ is zero. For a Gaussian envelope,

$$E(t)=E_{t=0}e^{-(t/\tau_{1/e})^2/2}\ , \tag{29}$$

we obtain

$$\theta(q_k)=\Theta e^{-(q_k/q_{1/e})^2/2}\ , \tag{30}$$

where we have defined

$$q_{1/e}=\frac{m}{\hbar k\tau_{1/e}}\ . \tag{31}$$

Setting $\cos[\theta(q_k)/2]=1$, $\sin[\theta(q_k)/2]=\theta(q_k)/2$, and $\phi(q_k)=0$ leads to

$$F_1(\vec{r})=\left[\frac{q_0}{\sqrt{\pi}}\right]^{3/2}e^{-q_0^2\vec{r}^2/2} \tag{32}$$

and

$$F_2(\vec{r})=\left[\frac{q_0}{\sqrt{\pi}}\right]\left[\frac{q_{00}^2/q_0}{\sqrt{\pi}}\right]^{1/2}$$

$$\times\exp[-\tfrac{1}{2}(q_0^2r_1^2+q_{00}^2r_k^2)]\ , \tag{33}$$

where

$$\frac{1}{q_{00}^2}=\frac{1}{q_0^2}+\frac{1}{q_{1/e}^2}\ . \tag{34}$$

Thus the ground-state wave packet is spherical and unchanged while the excited-state wave packet, having an amplitude contour

$$q_0^2r_1^2+q_{00}^2r_k^2=\text{const}\ , \tag{35}$$

is ellipsoidal. Its length-to-width ratio L/W is just

$$\frac{L}{W}=\frac{q_0}{q_{00}}\ . \tag{36}$$

In the short-pulse limit $q_{00}=q_0$ and $L/W=1$. For very long pulses, however, $q_{00}\approx q_{1/e}$ and this ratio becomes

$$\frac{L}{W}=\frac{\tau_{1/e}}{\tau_D}\ . \tag{37}$$

The elongation of the excited-state wave packet has the effect of increasing the time it takes for it to separate from the ground-state wave packet. This is precisely what would be expected on the basis of a Doppler dephasing argument.[3] Long pulses have a narrow frequency spectrum and can only excite those atoms whose Doppler-shifted resonances are contained in a correspondingly narrow bandwidth. But such atoms take a longer time to dephase and one obtains the same result as that derived above.

ANGLED-BEAM ECHO GENERATION

In the view of the billard-ball model, photon echoes are generated when two or more excitation pulses create wave packets which later collide. Long-pulse excitation generally produces elongated wave packets whose transverse extent is unchanged from what it would have been using short pulses. If in addition the excitation pulses are not parallel the wave packets may not collide head on and complete overlap will not be possible. The unchanged transverse wave-packet extent however means that the sensitivity to excitation pulse angling is also unchanged. This can be used to advantage as an unambiguous method for determining the character of an atomic velocity distribution.

Consider the case where two excitation pulses have frequency ω but areas Θ_1 and Θ_2 and k vectors $\vec{k}_1$ and $\vec{k}_2$. Let the angle β between $\vec{k}_1$ and $\vec{k}_2$ be so small that it manifests itself only when observing differences between $\vec{k}_1$ and $\vec{k}_2$. Except for some obvious relabeling we proceed as before.

We start with unexcited atoms. The effect of the first pulse is to transform

$$\Psi(t)=F_1(\vec{r})\,|\,1\rangle \tag{38}$$

for $t<0$, into

$$\Psi(t)=\cos\frac{\Theta_1}{2}F_{1\to 1}(\vec{r})\,|\,1\rangle$$

$$+i\sin\frac{\Theta_1}{2}e^{-i(wt-\vec{k}_1\cdot\vec{r})}$$

$$\times F_{1\to 2}\left[\vec{r}-\frac{\hbar\vec{k}_1}{m}t\right]|\,2\rangle\ , \tag{39}$$

valid until the second pulse is applied at $t=\tau$. After $t=\tau$ we have

$$\Psi(t)=\cos\left[\frac{\Theta_1}{2}\right]\left[\cos\left[\frac{\Theta_2}{2}\right]F_{1\to1\to1}(\vec{r})\,|1\rangle+i\sin\left[\frac{\Theta_2}{2}\right]e^{-i[\omega(t-\tau)-\vec{k}_2\cdot\vec{r}]}F_{1\to1\to2}\left[\vec{r}-\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]|2\rangle\right]$$
$$+i\sin\left[\frac{\Theta_1}{2}\right]e^{-i(\omega\tau-\vec{k}_1\cdot\vec{r})}\left[\cos\left[\frac{\Theta_2}{2}\right]e^{-i\omega(t-\tau)}F_{1\to2\to2}\left[\vec{r}-\frac{\hbar\vec{k}_1}{m}t\right]|2\rangle\right.$$
$$\left.+i\sin\left[\frac{\Theta_2}{2}\right]e^{-i\vec{k}_2\cdot\vec{r}}F_{1\to2\to1}\left[\vec{r}-\frac{\hbar\vec{k}_1}{m}t+\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]|1\rangle\right]. \quad (40)$$

The subscripts on F show the evolution of states to the present state (indicated by the rightmost index for each F). The second pulse forms $F_{1\to2\to1}$ through stimulated emission of a photon. We illustrate the effect of the excitation pulses in Fig. 1. At $t=0^-$ we show a single spherical wave packet indicating an atom in its ground state. At $t=0^+$ we show the elongated excited-state wave packet created by the first excitation pulse. This elongated wave packet recoils with velocity $\hbar\vec{k}_1/m$. At $t=\tau_-$ the wave packets have separated; the second excitation pulse, directed along $\vec{k}_2$, arrives. Its immediate effect is shown at $t=\tau_+$. At this point we draw only the newly created wave packets corresponding to $F_{1\to2\to1}$ and $F_{1\to1\to2}$. We omit the wave packets corresponding to $F_{1\to2\to2}$ and $F_{1\to1\to1}$ as they do not contribute to the echo formation process. $F_{1\to1\to2}$ recoils along $\vec{k}_2$ while $F_{1\to2\to1}$ recoils transversely along $\vec{k}_1-\vec{k}_2$. When overlap occurs at $t=2\tau$ it is incomplete, as indicated in Fig. 1. The echo is correspondingly degraded and radiated along $2\vec{k}_2-\vec{k}_1$, the wave vector of the rephased polarization density. At $t=3\tau$ we show the wave packets when they are again well separated.

We use Eq. (14) to calculate the echo amplitude,

$$\vec{E}_{\text{echo}}=\hat{e}k^2\frac{e^{-i\vec{k}\cdot\vec{R}}}{R}NP_{21}\int d\vec{r}\,e^{-i(2\vec{k}_2-\vec{k}_1-\vec{k})\cdot\vec{r}}\cos\frac{\Theta_1}{2}\sin\frac{\Theta_1}{2}\sin^2\frac{\Theta_2}{2}$$
$$\times F^*_{1\to1\to2}\left[\vec{r}-\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]F_{1\to2\to1}\left[\vec{r}-\frac{\hbar\vec{k}_1}{m}t+\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]. \quad (41)$$

Assuming the sample is so small that $\exp[-i(2\vec{k}_2-\vec{k}_1-\vec{k})\cdot\vec{r}]\simeq1$ throughout, the echo will be radiated along $2\vec{k}_2-\vec{k}_1$. In that direction the echo amplitude is

$$\vec{E}_{\text{echo}}=\hat{e}\cos\frac{\Theta_1}{2}\sin\frac{\Theta_1}{2}\sin^2\frac{\Theta_2}{2}k^2\frac{e^{-i\vec{k}\cdot\vec{R}}}{R}NP_{21}$$
$$\times\int d\vec{r}\,F^*_{1\to1\to2}\left[\vec{r}-\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]F_{1\to2\to1}\left[\vec{r}-\frac{\hbar\vec{k}_1}{m}t+\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]. \quad (42)$$

For $\hat{k}$ along $\vec{k}_1$ and β small

$$\vec{k}_1=\vec{k}\;,\quad \vec{k}_2=k\beta\hat{1}+\vec{k}\;, \quad (43)$$

and the wave-packet amplitudes may be written as

$$F_{1\to1\to2}\left[\vec{r}-\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]=\left[\frac{q_0}{\sqrt{\pi}}\right]\exp\left[-\tfrac{1}{2}q_0^2\left[r_\perp-\frac{\hbar k\beta}{m}(t-\tau)\right]^2\right]f_{1\to1\to2}\left[r_k-\frac{\hbar k}{m}(t-\tau)\right],$$
$$F_{1\to2\to1}\left[\vec{r}-\frac{\hbar\vec{k}_1}{m}t+\frac{\hbar\vec{k}_2}{m}(t-\tau)\right]=\left[\frac{q_0}{\sqrt{\pi}}\right]\exp\left[-\tfrac{1}{2}q_0^2\left[r_\perp+\frac{\hbar k\beta}{m}(t-\tau)\right]^2\right]f_{1\to2\to1}\left[r_k-\frac{\hbar k}{m}\tau\right]. \quad (44)$$

The wave-packet overlap is then

$$\int d\vec{r}\, F^*_{1\to1\to2} F_{1\to2\to1} = \exp\left[-\left[\frac{q_0 \hbar k \beta}{m}\right]^2 (t-\tau)^2\right] \times \int dr_k f^*_{1\to1\to2}\left[r_k - \frac{\hbar k}{m}(t-\tau)\right] f_{1\to2\to1}\left[r_k - \frac{\hbar k}{m}\tau\right] . \tag{45}$$

The exponential term on the right-hand side of Eq. (45) can be rewritten as

$$\exp\{-[(t-\tau)/\tau_{\text{eff}}]^2\} , \tag{46}$$

where

$$\tau_{\text{eff}} = \frac{1}{\beta}\frac{m}{q_0 \hbar k} = \frac{1}{\beta}\tau_D . \tag{47}$$

For experiments in which τ_{eff} is long compared to the time during which appreciable overlap is maintained between the ground and excited state, the integral

$$G(t-2\tau) = \int dr_k f_{1\to1\to2}\left[r_k - \frac{\hbar k}{m}(t-\tau)\right] \times f_{1\to2\to1}\left[r_k - \frac{\hbar k}{m}\tau\right] \tag{48}$$

in Eq. (45) is sharply peaked relative to the exponential factor and we can write

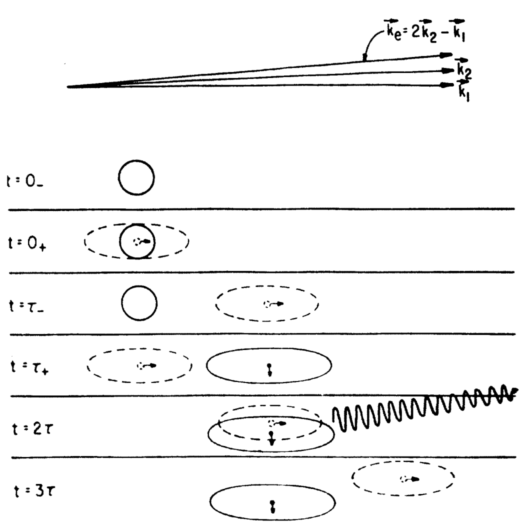

FIG. 1. Sequence of events leading to a photon echo using angled sub-Doppler-linewidth excitation pulses. Both pulses are assumed to be in the small-angle limit. The solid-line contour denotes the ground state and the dashed-line contour denotes the excited state.

$$\int d\vec{r}\, F^*_{1\to1\to2} F_{1\to2\to1} = e^{-(\tau/\tau_{\text{eff}})^2} G(t-2\tau) . \tag{49}$$

Thus, independently of the area or length of the excitation pulses, the echo amplitude falls off as a Gaussian in τ. If we work entirely in the small area limit with both pulses being Gaussian and having width $\tau_{1/e} \gg \tau_D$ then the ratios L/W for $F_{1\to1\to2}$ and $F_{1\to2\to1}$ are $\tau_{1/e}/\tau_D$ and $\sqrt{2}(\tau_{1/e}/\tau_D)$, respectively, and

$$G(t-2\tau) = G(0) \times \exp\{-[(t-2\tau)/\sqrt{3}\tau_{1/e}]^2/2\} . \tag{50}$$

THEORY

In this section we present a more general analysis of the billiard-ball echo problem. We show that the effective distribution functions $A_1(\vec{q})$ and $A_2(\vec{q})$ factor according to Eq. (11) and we obtain an explicit expression for the longitudinal distribution functions $a_1(q_k)$ and $a_2(q_k)$. We demonstrate that small area pulses excite atoms in proportion to the Fourier component of the radiation field at the atom's Doppler-shifted resonance frequency. We then consider the multiple-pulse excitation problem. Finally we justify our neglect of wave-packet spreading in the preceding sections.

We begin by considering an atom in its ground state and localized at the origin:

$$|\Psi\rangle = \int d\vec{q}\, A(\vec{q}) e^{-i(\hbar \vec{q}^2/2m)t} |\vec{q}\rangle |1\rangle . \tag{51}$$

We apply an excitation pulse of the form

$$E\left[t - \frac{r_k}{c}\right] \cos\left[\omega\left[t - \frac{r_k}{c}\right]\right] . \tag{52}$$

The appropriate Hamiltonian for this problem is

$$\hat{H} = \hat{H}_0 + \hat{H}_1 , \tag{53}$$

where

$$\hat{H}_0 = \frac{\hbar^2 \hat{\vec{q}}^2}{2m} + \hat{H}_{\text{internal}} , \tag{54}$$

$\hat{H}_{\text{internal}}$ being the atom's internal electronic Hamiltonian, and

$$\hat{H}_1 = -\hat{P}E\left[t-\frac{\hat{r}_k}{c}\right]\cos\left[\omega\left[t-\frac{\hat{r}_k}{c}\right]\right] \tag{55}$$

(the caret denotes quantities that are quantum-mechanical operators).

The Schrödinger equation that governs the time development of $|\Psi(t)\rangle$ is

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle = (\hat{H}_0+\hat{H}_1)|\Psi(t)\rangle . \tag{56}$$

We find it convenient to work in an interaction representation defined by

$$|\Phi(t)\rangle = e^{(i/\hbar)\hat{H}_0 t}|\Psi(t)\rangle . \tag{57}$$

The Schrödinger equation that governs the time development of $|\Phi(t)\rangle$ is

$$i\hbar\frac{\partial}{\partial t}|\Phi(t)\rangle = e^{(i/\hbar)\hat{H}_0 t}\hat{H}_1 e^{-(i/\hbar)\hat{H}_0 t}|\Phi(t)\rangle . \tag{58}$$

For times before and after the wave packet sees the pulse, $|\Phi(t)\rangle$ is time independent. We therefore write

$$|\Phi\rangle_{\text{after}} = \hat{U}\,|\Phi\rangle_{\text{before}} , \tag{59}$$

where $|\Phi\rangle_{\text{before}}$ is given by Eqs. (51) and (57) as

$$|\Phi\rangle_{\text{before}} = \int d\vec{q}\,A(\vec{q})\,|\vec{q}\rangle\,|1\rangle . \tag{60}$$

Equation (50) can be solved formally for $\hat{U}$, giving

$$\hat{U} = T\exp\left[-\frac{i}{\hbar}\int_{\text{before}}^{\text{after}} dt\, e^{(i/\hbar)\hat{H}_0 t} \times \hat{H}_1 e^{-(i/\hbar)\hat{H}_0 t}\right] , \tag{61}$$

where T indicates that the time-ordered product is to be used in the expansion of the exponential. When applying $\hat{U}$ to $|\Phi\rangle_{\text{before}}$ we can extend the limits of the integral over time in $\hat{U}$ to $\pm\infty$, since $\hat{H}_1$ is assumed to be zero in the region where $\langle\vec{r}\,|\Phi\rangle_{\text{before}}$ is nonzero for these extended times giving

$$\hat{U} = T\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{\infty} dt\, e^{(i/\hbar)\hat{H}_0 t} \times \hat{H}_1 e^{-(i/\hbar)\hat{H}_0 t}\right] . \tag{62}$$

We now make connection with Eq. (8) of the text by equating $|\Phi\rangle_{\text{after}}$ with $|\Psi(t=0^+)\rangle$,

$$\hat{U}\,|\Phi\rangle_{\text{before}} = \cos\left[\frac{\Theta}{2}\right]\int d\vec{q}\,A_1(\vec{q})\,|\vec{q}\rangle\,|1\rangle + i\sin\left[\frac{\Theta}{2}\right]\int d\vec{q}\,A_2(\vec{q})\,|\vec{q}+\vec{k}_{\vec{q}}\rangle\,|2\rangle . \tag{63}$$

We define $\vec{k}_q$ as the wave vector of a photon that can promote an atom initially in its ground state and having center-of-mass momentum $\vec{q}$ to its excited state having center-of-mass momentum $\vec{q}+\vec{k}_{\vec{q}}$. We will require the direction of $\vec{k}_{\vec{q}}$ to be the same as the direction of the wave vector of the applied laser pulse. Conservation of energy requires $\vec{k}_{\vec{q}}$ to satisfy the equation

$$c\,|\vec{k}_{\vec{q}}| + \frac{\hbar\vec{q}^{\,2}}{2m} = \Omega + \frac{\hbar(\vec{q}+\vec{k}_{\vec{q}})^2}{2m} . \tag{64}$$

We now invert Eq. (63) to find $A_1(\vec{q})$ and $A_2(\vec{q})$, giving

$$A_1(\vec{q}) = \frac{1}{\cos(\Theta/2)}\langle\vec{q}\,|\langle 1|\,\hat{U}\,|\Phi\rangle_{\text{before}} \tag{65}$$

and

$$A_2(\vec{q}) = \frac{1}{i\sin(\Theta/2)}\langle\vec{q}+\vec{k}_{\vec{q}}\,|\langle 2|\,\hat{U}\,|\Phi\rangle_{\text{before}} . \tag{66}$$

Writing $|\vec{q}\rangle = |q_k\rangle\,|q_\perp\rangle$ and $|\vec{q}+\vec{k}_{\vec{q}}\rangle = |q_k+k_{\vec{q}}\rangle\,|q_\perp\rangle$, Eqs. (65) and (66) give

$$A_1(\vec{q}) = \frac{1}{\cos(\Theta/2)}\frac{e^{-q_\perp^2/2q_0^2}}{(\pi q_0^2)^{3/4}}\left\langle q_k\left|\left\langle 1\left|\hat{U}\int dq_k'\,e^{-q_k'^2/2q_0^2}\right|q_k'\right\rangle\right|1\right\rangle \tag{67}$$

and

$$A_2(\vec{q}) = \frac{1}{i\sin(\Theta/2)}\frac{e^{-q_\perp^2/2q_0^2}}{(\pi q_0^2)^{3/4}}\left\langle q_k+k_{\vec{q}}\left|\left\langle 2\left|\hat{U}\int dq_k'\,e^{-q_k'^2/2q_0^2}\right|q_k'\right\rangle\right|1\right\rangle . \tag{68}$$

Comparing Eqs. (67) and (68) with Eq. (11) of the text we find

$$a_1(q_k) = \frac{1}{(\pi q_0^2)^{1/4}}\frac{1}{\cos(\Theta/2)}\left\langle q_k\left|\left\langle 1\left|\hat{U}\int dq_k'\,e^{-q_k'^2/2q_0^2}\right|q_k'\right\rangle\right|1\right\rangle \tag{69}$$

and

$$a_2(q_k)=\frac{1}{(\pi q_0^2)^{1/4}}\frac{1}{i\sin(\Theta/2)}\left\langle q_k+k_{\vec{q}}\left|\left\langle 2\left|\hat{U}\int dq_k' e^{-q_k'^2/2q_0^2}\right|q_k'\right\rangle\right|1\right\rangle . \tag{70}$$

These expressions are valid for pulses of arbitrary area.

We now find explicit expressions for $a_1(q_k)$ and $a_2(q_k)$ in the small-pulse-area limit. We proceed by expanding $\hat{U}$ and retaining only terms to the first order in $\hat{H}_1$. This gives

$$\hat{U}=1+\hat{U}_1 , \tag{71}$$

where

$$\hat{U}_1=-\frac{i}{\hbar}\int_{-\infty}^{\infty}dt\, e^{(i/\hbar)\hat{H}_0t}\hat{H}_1e^{-(i/\hbar)\hat{H}_0t} . \tag{72}$$

In this approximation Eq. (69) gives

$$a_1(q_k)=\frac{1}{(\pi q_0^2)^{1/4}}e^{-q_k^2/2q_0^2} , \tag{73}$$

where we have set $\cos(\Theta/2)=1$. Thus after the excitation pulse has passed, the ground-state wave packet is spherical and unchanged from what it was before the pulse, as expected. Equation (70) for $a_2(q_k)$ becomes, in the small-pulse-area limit,

$$a_2(q_k)=\frac{-1}{\hbar\frac{\Theta}{2}(\pi q_0^2)^{1/4}}\left\langle q_k+k_{\vec{q}}\left|\left\langle 2\left|\int_{-\infty}^{\infty}dt\, e^{(i/\hbar)\hat{H}_0t}\hat{H}_1e^{-(i/\hbar)\hat{H}_0t}\int dq_k' e^{-q_k'^2/2q_0^2}\right|q_k'\right\rangle\right|1\right\rangle \tag{74}$$

or

$$a_2(q_k)=\frac{P_{21}}{\hbar\Theta(\pi q_0^2)^{1/4}}\int dq_k' e^{-q_k'^2/2q_0^2}\int_{-\infty}^{\infty}dt\,\exp i\left[\Omega+\frac{\hbar(q_k+k_{\vec{q}})^2}{2m}-\frac{\hbar q_k'^2}{2m}\right]t$$
$$\times\left\langle q_k+k_{\vec{q}}\left|E\left[t-\frac{\hat{r}_k}{c}\right](e^{i\omega(t-\hat{r}_k/c)}+e^{-i\omega(t-\hat{r}_k/c)})\right|q_{k'}\right\rangle . \tag{75}$$

We now express $E(t-\hat{r}_k/c)$ in terms of its Fourier transform $\tilde{E}(u)$,

$$\tilde{E}(u)=\int_{-\infty}^{\infty}dt\,E(t)e^{-iut} . \tag{76}$$

This gives in Eq. (75)

$$a_2(q_k)=\frac{P_{21}}{2\pi\hbar\Theta(\pi q_0^2)^{1/4}}\int dq_k' e^{-q_k'/2q_0^2}\int_{-\infty}^{\infty}dt\int_{-\infty}^{\infty}du\,\tilde{E}(u)\exp i\left[ck_{\vec{q}}+\hbar\frac{q_k^2-q_k'^2}{2m}+u\right]t$$
$$\times\langle q_k+k_{\vec{q}}\,|\,(e^{i\omega t}e^{-i(\omega+u)\hat{r}_k/c}+e^{-i\omega t}e^{i(\omega-u)\hat{r}_k/c})\,|\,q_k'\rangle , \tag{77}$$

where we have used Eq. (64) to set

$$\Omega+\frac{\hbar(q_k+k_{\vec{q}})^2}{2m}=ck_{\vec{q}}+\frac{\hbar q_k^2}{2m}$$

in the first exponential term of Eq. (75). Noting that

$$\langle \vec{q}_1 | e^{i\vec{q}_2\cdot\hat{\vec{r}}} | \vec{q}_3 \rangle = \delta^{(3)}(\vec{q}_1-\vec{q}_2-\vec{q}_3)$$

we now write Eq. (77) as

$$a_2(q_k)=\frac{P_{21}}{2\pi\hbar\Theta(\pi q_0^2)^{1/4}}\int dq_k' e^{-q_k'^2/2q_0^2}\int_{-\infty}^{\infty}dt\int_{-\infty}^{\infty}du\,\tilde{E}(u)\exp i\left[ck_{\vec{q}}+\hbar\frac{q_k^2-q_k'^2}{2m}+u\right]t$$
$$\times\left[e^{i\omega t}\delta\left[q_k+k_{\vec{q}}+\frac{\omega}{c}+\frac{u}{c}-q_k'\right]\right.$$
$$\left.+e^{-i\omega t}\delta\left[q_k+k_{\vec{q}}-\frac{\omega}{c}+\frac{u}{c}-q_k'\right]\right]. \tag{78}$$

Evaluating the integrals over t and then over u gives

$$a_2(q_k)=\frac{P_{21}}{\hbar\Theta(\pi q_0^2)^{1/4}}\int dq_k' e^{-q_k'^2/2q_0^2}\left[\tilde{E}\left[-ck_{\vec{q}}-\omega-\hbar\frac{q_k^2-q_k'^2}{2m}\right]+\tilde{E}\left[-ck_{\vec{q}}+\omega-\hbar\frac{q_k^2-q_k'^2}{2m}\right]\right]$$
$$\times\left[\frac{\delta(q_k'-q_k)}{\left|-1+\dfrac{\hbar q_k}{mc}\right|}+\frac{\delta(q_k'-2mc/\hbar+q_k)}{\left|1-\dfrac{\hbar q_k}{mc}\right|}\right]. \tag{79}$$

Because of the $e^{-q_k'^2/2q_0^2}$ term in the integral the second delta function's contribution is negligible compared to that of the first. If we now make the approximation that $-1+q_k/m=-1$ then Eq. (79) gives

$$a_2(q_k)=\frac{P_{21}}{\hbar\Theta(\pi q_0^2)^{1/4}}e^{-q_k^2/2q_0^2}[\tilde{E}(-ck_{\vec{q}}-\omega)+\tilde{E}(-ck_{\vec{q}}+\omega)]\,. \tag{80}$$

For the excitation pulses under consideration $|\tilde{E}(-ck_{\vec{q}}+\omega)| \gg |\tilde{E}(-ck_{\vec{q}}-\omega)|$ so that this reduces to

$$a_2(q_k)=\frac{P_{21}}{\hbar\Theta(\pi q_0^2)^{1/4}}e^{-q_k^2/2q_0^2}\tilde{E}(\omega-ck_{\vec{q}})\,. \tag{81}$$

Relabeling $ck_{\vec{q}}$ as ω_{q_k}, the frequency of the Fourier component of the radiation field at the atom's Doppler-shifted resonance, and then using Eq. (27) for $\theta(q_k)$, Eq. (81) gives the simple result

$$a_2(q_k)=\frac{1}{(\pi q_0^2)^{1/4}}\frac{\theta(q_k)}{\Theta}e^{-q_k^2/2q_0^2}\,. \tag{82}$$

This justifies the use of Eq. (27) for the pulse area in the small-pulse limit.

For a Gaussian-shaped excitation pulse in the small-pulse-area limit, as given in Eq. (29),

$$\tilde{E}(\omega-ck_{\vec{q}})=\tilde{E}\left[-\frac{\hbar\vec{k}\cdot\vec{q}}{m}\right]$$
$$=2\tau_{1/e}\sqrt{\pi}E_{t=0}e^{-[\hbar q_k/(m/k\tau_{1/e})]^2/2} \tag{83}$$

and

$$a_2(q_k)=\frac{1}{(\pi q_0^2)^{1/4}}e^{-q_k^2/2q_{00}^2}\,, \tag{84}$$

where q_{00} is defined in Eq. (34). We summarize by saying that for a Gaussian pulse in the small-pulse-area limit the interaction picture wave function of an atom after the pulse has passed is given by

$$|\Phi\rangle_{\text{after}}=\int d\vec{q}A(\vec{q})|\vec{q}\rangle|1\rangle+i\frac{\Theta}{2}\int d\vec{q}A(\vec{q})e^{-[\hbar\vec{q}\cdot\vec{k}/(m/\tau_{1/e})]^2/2}|\vec{q}+\vec{k}_{\vec{q}}\rangle|2\rangle\,. \tag{85}$$

Equation (85) can now be generalized to multiple-pulse excitations. Consider an atom with an interaction picture wave function given initially by Eq. (60). After interacting with a series of N Gaussian-shaped laser pulses, all in the small-pulse-area limit, all having the same time width $\tau_{1/e}$ and having wave vectors $\vec{k}_1,\vec{k}_2,\ldots,\vec{k}_N$, the center-of-mass part of the wave function that has interacted with all the pulses will be given by

$$\int d\vec{q}\,A(\vec{q})\prod_{i=1}^{N}(e^{-[\hbar\vec{k}_i\cdot\vec{q}/(m/\tau_{1/e})]^2/2})\left|\vec{q}+\sum_{i=1}^{N}\epsilon_i\vec{k}_i\right\rangle, \tag{86}$$

where $\epsilon_i=+1$ or -1 if a photon has been absorbed or emitted, respectively.

Finally, we show that wave-packet spreading can be neglected when calculating the coherently radiated electric field. The general form for the wave function of an atom that has seen a coherent excitation pulse is

$$|\Psi(t)\rangle=\cos\frac{\Theta}{2}\int d\vec{q}\,A_1(\vec{q})e^{-i\hbar(\vec{q}^{\,2}/2m)t}|\vec{q}\rangle|1\rangle + i\sin\frac{\Theta}{2}\int d\vec{q}\,A_2(\vec{q})e^{-i\hbar[(\vec{q}+\vec{k}_{\vec{q}})^2/2m]t}e^{-i\Omega t}|\vec{q}+\vec{k}_{\vec{q}}\rangle|2\rangle . \tag{87}$$

The spatial form of $|\Psi(t)\rangle$ is

$$\Psi(\vec{r}_1t)=\frac{\cos\frac{\Theta}{2}}{(2\pi)^{3/2}}\int d\vec{q}\,A_1(\vec{q})e^{-i\hbar(\vec{q}^{\,2}/2m)t}e^{i\vec{q}\cdot\vec{r}}|1\rangle + i\frac{\sin\frac{\Theta}{2}}{(2\pi)^{3/2}}e^{-i\Omega t}\int d\vec{q}\,A_2(\vec{q})e^{-i\hbar[(\vec{q}+\vec{k}_{\vec{q}})^2/2m]t}e^{i(\vec{q}+\vec{k}_{\vec{q}})\cdot\vec{r}}|2\rangle . \tag{88}$$

The coherently radiated electric field is proportional to the matrix element[1]

$$\langle\Psi(t)|\hat{P}e^{i\vec{k}\cdot\vec{r}}|\Psi(t)\rangle . \tag{89}$$

If we call this matrix element J and retain only those terms that will remain after averaging over a macroscopic sample we are left with

$$J\propto\int d\vec{r}\left[\int d\vec{q}\,'A_2^*(\vec{q}\,')e^{i\hbar[(\vec{q}\,'+\vec{k}_{\vec{q}\,'})^2/2m]t}e^{i\Omega t}e^{-i(\vec{q}\,'+\vec{k}_{\vec{q}\,'})\cdot\vec{r}}\right]e^{i\vec{k}\cdot\vec{r}} \times\left[\int d\vec{q}\,A_1(\vec{q})e^{-i\hbar(\vec{q}^{\,2}/2m)t}e^{i\vec{q}\cdot\vec{r}}\right]. \tag{90}$$

Since $|\vec{k}_{\vec{q}}-\vec{k}|/q_0\ll1$ we can write

$$J\propto e^{i\omega t}\int d\vec{r}\left[\int d\vec{q}\,'A_2^*(\vec{q}\,')e^{i\hbar(\vec{q}\,'^2/2m)t}e^{-i\vec{q}\,'\cdot[\vec{r}-(\hbar\vec{k}/m)t]}\right] \times\left[\int d\vec{q}\,A_1(\vec{q})e^{-i\hbar(\vec{q}^{\,2}/2m)t}e^{i\vec{q}\cdot\vec{r}}\right], \tag{91}$$

where $\omega=\Omega+\hbar\vec{k}^{\,2}/2m$. The integral over $\vec{r}$ gives the delta function $(2\pi)^3\delta^{(3)}(\vec{q}-\vec{q}\,')$. When we do the $\vec{q}$ or $\vec{q}\,'$ integral, the term $e^{i\hbar(\vec{q}\,'^2/2m)t}$ which represents wave-packet spreading of the excited state cancels the term $e^{-i\hbar(\vec{q}^{\,2}/2m)t}$, which represents wave-packet spreading of the ground state. The wave-packet spreading terms can therefore be dropped in Eq. (91) giving

$$J\propto e^{i\omega t}\int d\vec{r}\left[\int d\vec{q}\,'A_2^*(\vec{q}\,')e^{i\vec{q}\,'\cdot[\vec{r}-(\hbar\vec{k}/m)t]}\right] \times\left[\int d\vec{q}\,A_1(\vec{q})e^{i\vec{q}\cdot\vec{r}}\right], \tag{92}$$

$$J\propto e^{i\omega t}\int d\vec{r}\,F_2^*\left[\vec{r}-\frac{\hbar\vec{k}}{m}t\right]F_1(\vec{r}) . \tag{93}$$

Therefore, the initial unspread wave packets can be used throughout when calculating the coherently radiated electric field; only the recoil of the wave packets due to absorbing or emitting a photon need be taken into account.

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