

PHYSICAL REVIEW LETTERS

VOLUME 84

24 APRIL 2000

NUMBER 17

Coherent Scattering with Pulsed Matter Beams

F. Robicheaux^{1,2} and L. D. Noordam²

¹*Department of Physics, Auburn University, Auburn, Alabama 36849*

²*FOM Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*
(Received 16 June 1999; revised manuscript received 11 January 2000)

We present a quantum theory for the interaction of a quantum target with a time dependent matter beam. When several pulses in the incident beam arrive with a period τ , transitions between levels with an energy difference h/τ can be enhanced. Unlike all previous studies, we find that transitions in passive targets can distinguish between an incoherent beam and a beam with a coherent wave packet structure. As an example, we calculate the transition probability of Rb Rydberg atoms interacting with a pulsed electron beam.

PACS numbers: 03.75.Fi, 03.65.Bz, 34.60.+z, 39.10.+j

Scattering between two material objects has served as an excellent probe of the processes important for chemical reactions and atomic, nuclear, and particle physics. In all of these studies, the incident beam has not contained any measurable type of longitudinal coherence [1]. An example of longitudinal coherence is when the wave function for an incident particle is a peaked wave packet along the beam. When longitudinal coherence is not important, the transition rate between two states in the target equals the cross section for the transition times the current density of incident particles. Also, each momentum component of a projectile can be treated independently; unlike a wave packet, the different momentum components have an unspecified phase relationship.

There have been some experimental [1–3] and theoretical [4–9] investigations into the effect of a coherent wave packet structure in the incident beam. While there has not yet been a scattering experiment that utilizes a longitudinal coherence from off-diagonal density matrix elements, $\rho(\mathbf{k}, \mathbf{k}')$, in the incident beam, the development of “atom lasers” [10–13] and the “pulsed electron gun” [14,15] seems to promise a source of coherent incident matter waves that will allow such a measurement. In the atom lasers, the matter waves have a very long coherence length compared to the de Broglie wave length of an atom in the beam. In the pulsed electron gun, electrons are emitted in bunches with a time between bunches measured in tens of picoseconds.

In this Letter, we derive the impact that an incident matter beam composed of several pulses will have on a quantum system. As an example, we calculate the effect that a series of electron pulses [14,15] with an energy of ~ 1 keV has on a Rb Rydberg state when the pulses arrive at the atom with a period of 10–100 ps. In Fig. 1, we present a schematic drawing of this situation where three sheets of electrons are moving past a Rydberg atom and cause a transition in the atom. This interesting system may be compared to an atom inside a plasma [16] where excitation by an individual charged particle or by the fluctuating microfield can both play a role; in a plasma, it is impossible

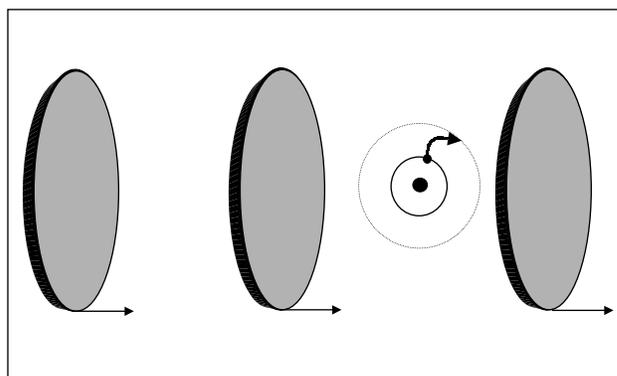


FIG. 1. A schematic drawing of a Rydberg atom interacting with an beam bunched in the longitudinal direction.

to control the fluctuating field, whereas in our system we can control which Fourier components of the quasiclassical electric field contribute to the transition. This system may also be contrasted with attempts to control reaction products through interaction of molecules or atoms with specially modulated light beams [17] *except* there have not been any proposals to control transitions by modulating a matter beam.

Intuitively, the incident beam in Fig. 1 causes transitions in the quantum target from two qualitatively different mechanisms. The first mechanism is the scattering of an individual electron by the quantum target. This gives a transition probability equal to the time integrated electron current density ζ times the inelastic cross section [18]. The second mechanism is the interaction of the Rydberg atom with the electric field generated by the charge density of the electron beam. This qualitative picture disagrees with previous theoretical investigations [6,7] which found that scattering from a passive target depends only on diagonal elements of the density matrix $\rho(\mathbf{k}, \mathbf{k})$, and thus coherence properties are irrelevant; off-diagonal elements are observed only when the beam interacts with a time dependent system like a vibrating mirror (see the experiment of Ref. [3]) or a target in a wave packet state. In Fig. 1, the electric field generated by the charge density (thus the transition probability) depends on off-diagonal elements $\rho(\mathbf{k}, \mathbf{k}')$ of the electron beam. We go beyond the usual treatment of transition probabilities where the effect of only one projectile is considered. We find that the qualitative picture is correct; transitions are driven by (i) the fluctuations in the incident beam or (ii) the temporal variation of the smooth classical-type field.

The theoretical development in this Letter is restricted to the situation where the quantum targets do not seriously attenuate the incident beam [19]. A second restriction is that only one incident object causes a transition in the target. Thus, we ignore high current density effects from one incident particle causing a transition $a \rightarrow b$ and then a second incident particle causing $b \rightarrow c$.

We treat the scattering as if the incident particles are completely coherent and can be treated as a normalizable wave packet of finite extent; this allows us to use a wave function description of the dynamics instead of a density matrix formalism. Incoherence in the incident particle beam is included by averaging over parameters in the wave packet. The incident particle is initially traveling in the z direction. For one incident particle and one scatterer, the normalized wave function is

$$\Psi(t) = \Phi_a e^{-iE_a t} \psi_a^{\text{inc}}(\mathbf{r}, t) + \sum_b \Phi_b e^{-iE_b t} \psi_{b \leftarrow a}^{\text{sca}}(\mathbf{r}, t), \quad (1)$$

where E_a is the energy and Φ_a is the wave function for state a of the target and $\psi_a^{\text{inc}}(\mathbf{r}, t)$ is the incident particle's wave function and $\psi_{b \leftarrow a}^{\text{sca}}(\mathbf{r}, t)$ is the scattered part of the wave function leaving the target in state b . Atomic units

are used unless explicitly stated otherwise. If the incident wave is delayed by an amount τ , the scattered wave is also delayed by an amount τ and it gets multiplied by $\exp[i(E_b - E_a)\tau]$; this phase change in the scattered wave plays a vital role for coherent scattering.

The probability for the incident particle to cause a transition in the target to state b is $P_{b \leftarrow a} = \langle \psi_{b \leftarrow a}^{\text{sca}} | \psi_{b \leftarrow a}^{\text{sca}} \rangle$. The longitudinal coherence of the incident wave (e.g., bunching the incident particles as in Fig. 1) has no measurable effect on the probability for causing a transition from state a to state b [6]. The probability for causing a transition is proportional to the inelastic cross section averaged over the energy distribution of the incident particle. Thus, the probability for causing a transition does not depend on the relative phases of the different momentum components, and thus whether or not the incident particle is in a wave packet or incoherent superposition of momentum components is irrelevant.

The analysis changes if there are N particles in the beam of transverse area $L_x L_y$. The transition probability from state a to b is usually assumed to be $N \langle \sigma_{b \leftarrow a} \rangle / L_x L_y \equiv \zeta \langle \sigma_{b \leftarrow a} \rangle$; i.e., the transition probability equals the inelastic cross section averaged over the momentum distribution in the incident beam times the time integral of the particle current density. This result arises from the assumption that each incident particle contributes incoherently to the transition. In this Letter, we demonstrate that if the packets for different incident particles arrive at the target in a correlated manner, then this assumption can be incorrect. To show this, we write out the N projectile wave function under the assumption that only one projectile causes a transition:

$$\begin{aligned} \Psi(t) = & \Phi_a e^{-iE_a t} \prod_{j=1}^N \psi_a^{\text{inc},j}(\mathbf{r}_j, [t - \tau_j]) \\ & + \sum_b \Phi_b e^{-iE_b t} \sum_{j'} \left\{ \psi_{b \leftarrow a}^{\text{sca},j'}(\mathbf{r}_{j'}, [t - \tau_{j'}]) e^{i\omega_{ba}\tau_{j'}} \right. \\ & \left. \times \prod_{j \neq j'} \psi_a^{\text{inc},j}(\mathbf{r}_j, [t - \tau_j]) \right\}, \quad (2) \end{aligned}$$

where $\omega_{ba} = E_b - E_a$ and the packet for the j th particle has been delayed by an amount τ_j . The j superscript on the incident and scattered wave function is meant to indicate that the wave packet for each incident particle is not necessarily related to any of the other packets. In Eq. (2), we made the assumption that the initial state of the incident beam is such that the wave function for the incident particles is a product of one particle functions. This situation can occur when the incident wave is the output from an atom laser since the atoms are bosons. This situation also holds when all lengths of a packet are smaller than the average distance between adjacent objects because the incident particles are distinguishable.

The probability for exciting the target to state b is

$$P_{b \leftarrow a}^{(N)} = \sum_j \langle \psi_{b \leftarrow a}^{\text{sca},j} | \psi_{b \leftarrow a}^{\text{sca},j} \rangle + \sum_{j \neq j'} \langle \psi_a^{\text{inc},j} | \psi_{b \leftarrow a}^{\text{sca},j} \rangle e^{i\omega_{ba}(\tau_j - \tau_{j'})} \langle \psi_{b \leftarrow a}^{\text{sca},j'} | \psi_a^{\text{inc},j'} \rangle \quad (3)$$

where unit normalization of the incident packets has been used. The first term of Eq. (3) is the incoherent sum of probabilities from each individual projectile, and the second term arises from the coherent effect of the projectiles on the target. It is important to remember that the coherent term is zero unless the incident wave packet has an energy width that is larger than the energy change in the target; if the energy width of the packet is too small there is no overlap between the incident and scattered waves because they do not contain the same energy components. As discussed below, the second term of Eq. (3) is proportional to off-diagonal elements of the two particle density matrix.

The scattered packet does not strongly overlap the incident packet because the incident packet has a momentum distribution strongly peaked in the z direction, whereas the scattered wave has a larger angular distribution of momentum. This means that $\langle \psi_{b \leftarrow a}^{\text{sca},j} | \psi_{b \leftarrow a}^{\text{sca},j} \rangle \gg |\langle \psi_a^{\text{inc},j} | \psi_{b \leftarrow a}^{\text{sca},j} \rangle|^2$. It is illustrative to use this fact to approximate Eq. (3) in the form

$$P_{b \leftarrow a}^{(N)} \approx \sum_{j=1}^N \langle \psi_{b \leftarrow a}^{\text{sca},j} | \psi_{b \leftarrow a}^{\text{sca},j} \rangle + \left| \sum_{j=1}^N \langle \psi_a^{\text{inc},j} | \psi_{b \leftarrow a}^{\text{sca},j} \rangle e^{i\omega_{ba}\tau_j} \right|^2, \quad (4)$$

which can serve as the basis for discussing the physical processes important for scattering with a pulsed incident beam. Although an individual contribution to the incoherent term is larger than one for the coherent term, there are N times more contributions to the coherent term. Therefore, the coherent contribution to the probability can be dominant for large numbers of projectiles N . We interpret the second term in Eq. (4) as arising from the coherent field from all of the projectiles acting on the target. This interpretation arises from the form of this term in which the amplitudes from each individual particle are superposed and the probability is the absolute value squared. Another reason for this interpretation is that in the first order Born approximation the second term in Eq. (4) *exactly* equals the transition from state a to state b calculated using first order time dependent perturbation theory and the time dependent coupling potential generated by the incident wave packets $|\psi_a^{\text{inc},j}|^2$.

In general, Eq. (4) can be quite complicated. But under the situations that hold in most experiments, this equation can be simplified and given in terms of familiar scattering parameters. The only extra condition is that the transverse length of the beam be large compared to the distance over which the target and the beam interact. We will also assume that the total cross section and the dif-

ferential cross section in the forward scattering direction do not vary strongly with energy over the energy width of the wave packet. Under these circumstances Eq. (4) can be rewritten as

$$P_{b \leftarrow a}^{(N)} = \zeta \sigma_{b \leftarrow a} + \zeta^2 2\pi \left| \frac{1}{N} \sum_{j=1}^N e^{i\omega_{ba}\tau_j} \right|^2 \left(\frac{d\sigma_{b \leftarrow a}}{dq dq} \right) \Big|_{q=q_0}, \quad (5)$$

where q_0 is the minimum of the absolute value of the momentum transfer which occurs when the scattered wave travels in the same direction as the incident wave, i.e., $\theta = 0$; $q_0 \approx |\omega_{ba}|/k_0$ when the incident energy is large compared to the energy given to the target. The differential cross section $d\sigma/qdq = \int d\phi (d\sigma/d\Omega)/k_0 k_f$ with the k_0, k_f the initial and final wave numbers.

The incoherent contribution to the probability is the total cross section times the time integrated current density of incident particles just as expected. The coherent term in Eq. (5) is proportional to the differential cross section in the forward direction and, thus, becomes easier to measure for transitions where the cross section is strongly peaked in the forward direction. Note from Eq. (5) that the differential cross section in the forward direction can be obtained by measuring the *total* transition probability as a function of ζ . The coherent contribution to the transition probability can be as large as $2\pi\zeta^2 d\sigma/qdq$. The minimum time integrated current density necessary to have the coherent term equal the incoherent term is $\zeta_{\text{min}} = (2\pi)^{-1} \sigma_{b \leftarrow a} / (d\sigma_{b \leftarrow a} / dqdq)$; at this value, the incoherent contribution to the probability is $(2\pi)^{-1} \sigma_{b \leftarrow a}^2 / (d\sigma_{b \leftarrow a} / dqdq)$. The incoherent probability $\sigma_{b \leftarrow a} \zeta_{\text{min}}$ must be much less than 1 for the coherence effects to be measurable.

We want to draw attention to the general features of this derivation. In *all* situations, a collimated beam of particles interacting with a quantum target in an eigenstate a can cause transitions to state b through two mechanisms. The first is proportional to $\int \rho(\mathbf{k}, \mathbf{k}) \sigma_{b \leftarrow a} d^3 \mathbf{k}$ and is the usual incoherent scattering; this term depends only on diagonal elements of the density matrix and thus cannot manifest a difference between an incoherent beam and a beam with a coherent wave packet structure. The second is a coherent scattering term which is proportional to $\int \rho(\mathbf{k} + q_0 \hat{z}, \mathbf{k}'; \mathbf{k}, \mathbf{k}' + q_0' \hat{z}) T_{b \leftarrow a}(q_0) T_{b \leftarrow a}^*(q_0') dk_z dk_z'$, where T is the inelastic transition amplitude in the forward direction and ρ is the two particle density matrix at $t = 0$ [for the case discussed above $\rho(\mathbf{k} + q_0 \hat{z}, \mathbf{k}'; \mathbf{k}, \mathbf{k}' + q_0' \hat{z}) = \rho(\mathbf{k} + q_0 \hat{z}, \mathbf{k}) \rho(\mathbf{k}', \mathbf{k}' + q_0' \hat{z})$]; this term has been ignored in previous studies either because most beams are incoherent or because the scattered wave is not sufficiently peaked in the forward direction. Because the second mechanism depends on the off-diagonal density matrix element, some types of longitudinal coherence will have an effect on this term.

As an example, we examined the effect of multiple electron pulse scattering on the $46s$ state of Rb. In all calculations we used a 1 keV incident electron beam to cause the transition; the first order Born approximation was used to calculate the scattering parameters. For the results presented here, we had 9 equally spaced electron pulses interact with the atom and examined the transition probability versus the time between the pulses. To compare the results for different values of ζ we plotted the probability for the transition divided by ζ ; this gives a parameter whose constant value equals the total cross section for the transition. In Fig. 2, we present the results from calculations for the transition from the $46s$ to the $45p$ states. For this transition, the incoherent scattering [first term Eq. (5)] populates all m components of the $45p$ state, whereas the coherent scattering [second term Eq. (5)] does not change J_z because the electron continues in the z direction after scattering for this term. The period of the transition is 23 ps, the cross section is $80 \text{ Tb} = 8 \times 10^{-11} \text{ cm}^2$, and the value of $\zeta_{\min} = 2.1 \times 10^4 \text{ cm}^{-2}$ which gives an incoherent probability of 1.7×10^{-6} . Note that such integrated current densities are well within experimental reach [20].

There is no effect from the coherent scattering in the $\zeta \rightarrow 0$ limit; the probability divided by ζ goes to the usual definition of the inelastic cross section in this limit. The coherent scattering is small unless the time between the electron pulses is a multiple of the transition period; as in driving a transition with a periodic electric field, the transition does not occur unless the period of the driving field is a multiple of the transition period. The relative size of the coherent scattering compared to the incoherent scattering increases linearly with the integrated particle current density of the beam. In this figure, it is easy to see the effect

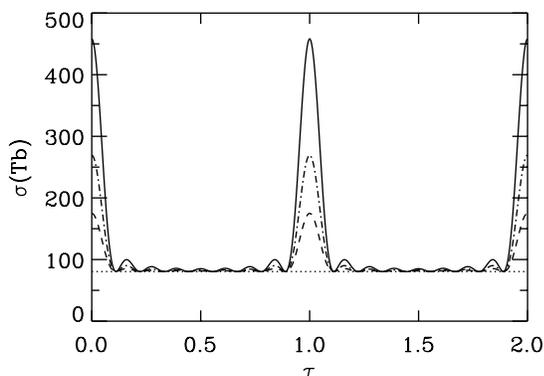


FIG. 2. Nine equally spaced electron pulses of 1 keV incident energy cause the transition from the $46s$ state of Rb. The time between pulses τ is given in units of the transition period. The transition probability is divided by ζ to give a cross section. The dotted line is the low current density, $\zeta \rightarrow 0$, result; i.e., the usual definition of cross section. The peaks at integer values of τ arise from the coherent scattering by the equally spaced electron pulses. The period for the $46s$ - $45p$ transition is 23 ps. $\zeta = 2.5, 5.0,$ and $10.0 \times 10^4 \text{ cm}^{-2}$ for the dashed, dot-dashed, and solid lines, respectively.

from the coherent pulses for the dipole allowed transitions. However, to observe a similar effect for nondipole transitions in Rydberg atoms will be difficult because the dipole allowed transitions are strongly peaked in the forward direction, whereas all other multipole transitions have a minimum in the forward scattering direction. A different way of interpreting the results is that the time varying electric field from the coherent excitation causes much stronger interactions between dipole allowed transitions than between nondipole allowed transitions.

There are some special features of the excitation scheme that have an effect on the excitation probability as a function of the time between pulses. The width in τ of the peaks depends on the number of pulses in the incident beam: the greater the number of pulses, the smaller the width. The height of the coherent peak only depends on ζ . Under most circumstances, ζ increases with the number of pulses in the beam; therefore, we expect to increase the effect of the coherent term relative to the incoherent term by increasing the number of pulses.

In conclusion, we have shown that it is possible to detect longitudinal aspects of coherence in an incident particle beam without using a time dependent detector [21]. We have found that there exists a coherent scattering term that depends on the time integrated current densities of incident particles. The parameters that control whether coherent or incoherent scattering dominates have been related to the total cross section and the differential cross section in the forward direction. The theory described in this paper suggests that control over the final state distribution can be achieved in a scattering experiment when the electron current is pulsed: transitions matching the pulse repetition period are strongly favored. Calculations for keV scattering of electrons on Rydberg states of Rb indicate that the particle current densities that will be needed to observe the coherent scattering are within the range of existing technology.

It is a pleasure to acknowledge discussions with R. B. Vrijen, H. G. Muller, K. B. MacAdam, and A. Tip. This work is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie (FOM)," which is financially supported by the "Nederlandse organisatie voor Wetenschappelijke Onderzoek (NWO)." F. R. is also supported by the NSF.

- [1] R. A. Rubenstein *et al.*, Phys. Rev. Lett. **82**, 2018 (1999); after submission of our paper, R. A. Rubenstein *et al.*, Phys. Rev. Lett. **83**, 2285 (1999) presented measurements of the longitudinal part of the density matrix of a beam.
- [2] H. Kaiser, S. A. Werner, and E. A. George, Phys. Rev. Lett. **50**, 560 (1983); **51**, 1106 (1983).
- [3] P. Szriftgiser *et al.*, Phys. Rev. Lett. **77**, 4 (1996).
- [4] A. G. Klein, G. I. Opat, and W. A. Hamilton, Phys. Rev. Lett. **50**, 563 (1983).
- [5] G. Comsa, Phys. Rev. Lett. **51**, 1105 (1983).

- [6] H. J. Bernstein and F. E. Low, *Phys. Rev. Lett.* **59**, 951 (1987).
- [7] R. Golub and S. K. Lamoreaux, *Phys. Lett. A* **162**, 122 (1992).
- [8] M. Raymer, *J. Mod. Opt.* **44**, 2565 (1997).
- [9] D. Kokorowski and D. Pritchard, *J. Mod. Opt.* **44**, 2575 (1997); A. Dhirani *et al.*, *J. Mod. Opt.* **44**, 2583 (1997).
- [10] M. R. Andrews *et al.*, *Science* **275**, 637 (1997).
- [11] M.-O. Mewes *et al.*, *Phys. Rev. Lett.* **78**, 582 (1997).
- [12] I. Bloch, T. W. Hänsch, and T. Esslinger, *Phys. Rev. Lett.* **82**, 3008 (1999).
- [13] E. W. Hagley *et al.*, *Science* **283**, 1706 (1999).
- [14] F. Robicheaux, G. M. Lankhuijzen, and L. D. Noordam, *J. Opt. Soc. Am. B* **15**, 1–5 (1998).
- [15] C. W. Rella *et al.*, *J. Opt. Soc. Am. B* **16**, 182–187 (1999).
- [16] M. S. Murillo and J. C. Weisheit, *Phys. Rep.* **302**, 1 (1998).
- [17] A. Assion *et al.*, *Science* **283**, 919 (1998).
- [18] R. G. Rolfes *et al.*, *J. Phys. B* **26**, 2191 (1993).
- [19] Using typical cross sections (less than 100 Tb = 10^{-10} cm²), Rydberg atom densities (less than 10^8 cm⁻³), and travel length (less than 1 cm), we find the probability for an electron to scatter is less than 1%.
- [20] D. Bisero *et al.*, *J. Appl. Phys.* **82**, 1384 (1997).
- [21] We find that there is the possibility for resonant excitation even in the $\zeta \rightarrow 0$ limit when the Rydberg atom is prepared in a wave packet. These investigations will be presented elsewhere.