Diffraction of Rydberg Atoms by Laser Light

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It is shown that semiclassical path representations provide a well-suited framework for the description of the diffraction of Rydberg atoms by laser light. They allow one to relate the momentum exchange between the atomic center-of-mass motion and the laser field directly to elementary laser-assisted collisions between the excited Rydberg electron and the ionic core. Thus besides a convenient calculational tool they provide also direct physical insight into the underlying internal dynamics of the Rydberg atoms.

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Deflection of atoms by laser light has been a problem of atomic physics of considerable experimental and theoretical interest during the last few years [1, 2]. So far investigations have concentrated mainly on two- and few-level systems as far as the internal dynamics of the atoms is concerned [3]. In this Letter effects originating from the coherent excitation of a large number of atomic Rydberg states by a standing-wave laser field are discussed.

Neglecting the atomic center-of-mass motion it has been shown recently [4] that laser-induced processes in which a large number of atomic Rydberg states close to a threshold are excited coherently from an energetically low-lying bound state lead to the preparation of a nonstationary, spatially localized electronic wave packet. As Rydberg states with low values of the angular momentum participate in such an excitation process it is only the radial coordinate of the excited Rydberg electron which is well localized in comparison with the large extension of typical Rydberg states. These excitation processes can be analyzed in a convenient way with the help of *semiclassical path representations* [4]. Thereby the relevant atomic transition amplitudes are related directly to (1) the quantum mechanical aspects of the atom-laser interaction which is localized typically in a reaction zone of a few Bohr radii around the atomic nucleus and (2) the classical paths of the excited Rydberg electron relative to the atomic nucleus. These semiclassical path representations may be obtained in the semiclassical limit from the corresponding Feynman path integral representations. Taking into account the atomic center-of-mass motion in the following it is shown that they also provide a particularly convenient framework for the description of the diffraction of Rydberg atoms by laser light.

A typical layout of a diffraction experiment is schematically shown in Fig. 1. A beam of atoms which are initially prepared in an energetically low-lying bound state $|g\rangle$ crosses a standing-wave laser beam at right angle. During the passage through the interaction region the atoms are excited to Rydberg states $|n\rangle$ by the laser field. For simplicity, we assume that the laser field may be approximated by $\mathbf{E}(\mathbf{x},t) = 2\mathcal{E}\Theta(z)\Theta(L-z)\sin(kx)\cos(\omega t)$ and we neglect ionization and spontaneous emission of photons from the excited Rydberg states. Thereby $\mathcal{E}$ is the laser polarization vector and $\Theta(z)$ is the unit step function. Furthermore, we assume that the kinetic energy of the atoms is much larger than the relevant interaction energy with the laser field and that during the passage through the interaction region the atomic motion in the direction of the standing wave can be neglected, i.e., $kv_x\tau << 1$. Under these conditions the center-of-mass motion of an atom through the interaction region can be described classically with a straight line trajectory [1]. This implies that the interaction time τ is related to the initial velocity $v_{\rm in} = P_{\rm in}/M$ and the beam diameter L by $\tau = L/v_{\rm in}$ and that the standing wave leads to a periodic modulation of the relevant quantum mechanical transition amplitudes.

In this approximation the semiclassical path representation for the probability amplitude of detecting an atom after passage through the interaction region in the initial state $|g\rangle$ with momentum $P_x = \pm 2lk$ $(l = 0, 1, \ldots)$ is given by [1]

$$a_g^{(l)}(\tau) = \frac{k}{2\pi}\int_0^{2\pi/k} dx\, e^{-2ilkx} a_g(x,\tau) e^{-i[\epsilon_g + P_{\rm in}^2/2M]\tau} \tag{1}$$

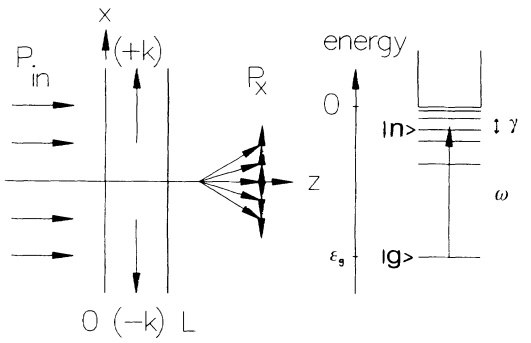

FIG. 1. Diffraction of an atomic beam by a standing-wave laser field and internal excitation process.

with [4]

$$a_g(x,\tau) = e^{-i(\delta\omega - i\gamma/2)\sin^2(kx)\tau} + \sum_{M=0}^{\infty} \int_{-\infty}^{0} d\epsilon\, e^{-i(\epsilon-\omega)\tau} [\gamma/(2\pi)][\epsilon - \bar{\epsilon} - (\delta\omega - i\gamma/2)\sin^2(kx)]^{-2} \sin^2(kx) e^{2i\pi(\nu+\alpha)} [\tilde{\chi} e^{2i\pi\nu}]^M \tag{2}$$

and $\epsilon = -1/(2\nu)^2$ (hartree atomic units are used). The mean excited energy is denoted $\bar{\epsilon} = \epsilon_g + \omega$. Within this model atoms in state $|\, g\rangle$ are only found with momenta $P_x = \pm 2lk$, whereas atoms in excited states $|\, n\rangle$ have momenta $P_x = \pm 2(l+1)k$ [1]. Physically this is due to the fact that for the observation of an atom in the initial state, for example, after the interaction with the laser field an equal number of photon absorption and stimulated emission events is required each of which transfers a momentum of $\Delta P_x = \pm k$ to the center of mass of the atom. The first term in Eq. (2) describes the depletion of the initial state $|\, g\rangle$ by the laser field according to an exponential decay. The Mth term in the sum is the probability amplitude of the Mth return of the excited Rydberg electron to the core region. The quantity $2\pi\nu$ is the classical action of a Rydberg electron of energy ϵ which moves along a classical periodic Coulomb orbit with zero angular momentum [4].

The quadratic Stark shift $\delta\omega$ and the ionization rate γ (both evaluated at maximum field strength) characterize the atom-laser interaction. In particular, the ionization rate γ determines the excited energy range. The quadratic Stark shift includes the ponderomotive energy shift of the excited Rydberg states. The scattering matrix element

$$\tilde{\chi} = \chi - e^{2i\pi\alpha} i\gamma \sin^2(kx)[\epsilon - \bar{\epsilon} - (\delta\omega - i\gamma/2)\sin^2(kx)]^{-1}$$

describes laser-assisted electron-ion scattering of the excited Rydberg electron from the core region [4]. The electron-ion scattering matrix element χ describes the scattering of the excited Rydberg electron by the residual atomic electrons inside the ionic core [5]. In the simple case of an alkali atom with one valence electron, which we consider here, it is given by $\chi = e^{2i\pi\alpha}$ with the quantum defect α. All these quantities are slowly varying functions of energy across the photoionization threshold. This reflects the fact that the atom-laser interaction and electron-correlation effects are localized in a reaction zone which typically extends only a few Bohr radii around the atomic nucleus [4].

Equation (1) can be simplified further if the laser field excites Rydberg states sufficiently far below threshold in the sense that $|\, \bar{\epsilon}\, | >> \gamma, |\, \delta\omega\, |$ and if the excited energy range is sufficiently small so that dispersion is negligible, i.e., $M/2(d^2\nu/d\epsilon^2) << \gamma^{-2}, |\, \delta\omega\, |^{-2}$. In this case Eq. (1) reduces to

$$a_g^{(l)}(\tau) = I_l(\tau) + \sum_{M=1}^{\infty} \Theta(\tau - MT)[e^{2i\pi(\overline{\nu}+\alpha)}]^M \times \sum_{r=0}^{M-1} \binom{M-1}{r} \frac{[\gamma(\tau - MT)]^{r+1}}{(r+1)!} 4^{-(r+1)} \sum_{\mu=0}^{2(r+1)} \binom{2(r+1)}{\mu} (-1)^{\mu} I_{l+r+1-\mu}(\tau - MT) \tag{3}$$

with

$$I_l(t) = e^{-i[\delta\omega - i\gamma/2]t/2} e^{il\pi/2} J_l([\delta\omega - i\gamma/2]t/2) \tag{4}$$

and the Bessel function $J_l(x)$ of integer order. The mean classical orbit time of the excited Rydberg electron is denoted $T = 2\pi\overline{\nu}$ and $\overline{\nu}$ is its mean principal quantum number. The semiclassical path representation of Eq. (3) is the central result of this Letter and gives a uniform description of the state-selective atomic scattering amplitude. It is valid in the two-level limit where only one Rydberg state is excited significantly, i.e., $\gamma T << 1$, as well as in cases where many Rydberg states are excited coherently, i.e., $\gamma T >> 1$. Furthermore, it allows for a straightforward interpretation of the atomic scattering process in terms of probability amplitudes associated with repeated laser-assisted collisions between the excited Rydberg electron and the ionic core during the flight of the atom through the standing-wave laser field.

The first term in Eq. (3), i.e., $I_l(\tau)$, describes the momentum exchange between atom and laser field during the depletion of the initial atomic state $|\, g\rangle$. For interaction times $\tau < T$ it is the only contribution to $a_g^{(l)}(\tau)$. In the special case of a negligible quadratic Stark shift the quantity $I_l(\tau)$ reduces to the probability distribution of a classical unbounded symmetrical random walk with the transition rate $(\gamma/8)$ [6].

If $\tau > T$ the excited Rydberg electron can return to the reaction zone during the motion of the atom through the laser field. With each crossing of the reaction zone the laser-induced coupling between initial and excited states is turned on and off again. Thus by stimulated emission from the excited Rydberg states and subsequent absorption two laser photons are exchanged during each of these laser-assisted electron-ion collisions inside the reaction zone. Consequently, the momentum of the center-of-mass motion of the atom is changed by an amount $\Delta P_x = 0$ or $\Delta P_x = \pm 2k$ because these photons can have the same or opposite momenta. The (M,r)th term in the sum of Eq. (3) is the probability amplitude of the Mth

return of the electron to the reaction zone with r intermediate laser-assisted electron-ion scatterings. Because of the $\Theta(x)$ this term only contributes for interaction times $\tau > MT$. The binomial coefficient $\binom{M-1}{r}$ is equal to the number of ways in which r such scattering events can be chosen out of a total number of $M-1$ intermediate returns of the electron to the reaction zone. The factor $[\gamma(\tau - MT)]^{r+1}$ implies that with each of these scatterings the excited Rydberg electron experiences a time delay. Similarly, $\binom{2(r+1)}{\mu}$ is the number of ways in which μ photons with momentum $+k$ (or $-k$) can be selected out from the total number of $2(r+1)$ photons which can be exchanged during $r+1$ crossings of the reaction zone.

Equation (3) clearly exhibits the main difference between coherent laser-induced excitation of many Rydberg states, i.e., $\gamma T >> 1$, and the two-level limit, i.e., $\gamma T << 1$. In the first case the probability amplitudes associated with different returns of the electron to the reaction zone are well separated in time and for a given interaction time typically only a few terms with low values of M and r contribute. In the two-level limit contributions associated with different electron-core collisions overlap in time so that they interfere quantum mechanically. In this case Eq. (3) reduces to the well-known results for a two-level system [1].

In Fig. 2 the probability $\mid a_g^{(l)}(\tau) \mid^2$ is shown for a case where the initial state is depleted on a time scale short in comparison with the mean classical orbit time. Thus a radial electronic Rydberg wave packet is generated by the laser-induced excitation process [4]. This implies that for times $\tau = L/v_{\text{in}} < T$ the initial state probability decays exponentially. At multiples of the mean classical orbit time T this wave packet returns to the reaction zone. Therefore for interaction times $\tau \approx MT$ the initial state probability is increased by stimulated transitions to the initial state. These recombination processes lead to narrow peaks of $\mid a_g^{(l)}(\tau) \mid^2$ whose widths are of the order of $1/\gamma$. As is also apparent from Fig. 2 for the detection of atoms in state $\mid g\rangle$ with momenta $P_x = 2lk$ $(l > 2)$ the interaction time has to be sufficiently long so that at least $l-2$ laser-assisted collisions between the excited Rydberg wave packet and the ionic core are possible.

Compared with conventional approaches which are based on dressed-state analysis [3] the semiclassical path representation discussed here offers the conceptual advantage of relating relevant quantum mechanical transition amplitudes directly to elementary laser-assisted electron-ion collision processes which lead to the exchange of momentum between the laser field and the atoms. Thus atomic diffraction by laser light can be used for studying the dynamics of atomic Rydberg wave packets. In addition, the sensitivity of state-selective transition probabilities with respect to the interaction time might be of practical relevance for velocity and state-selective preparation of atoms. This approach offers the possibility to include electron-correlation effects (coupling between Rydberg series and autoionization) [4], effects of weak external static fields [7] as well as effects of atomic motion along the standing wave [8], in a natural way. Therefore in the future it might become a valuable tool for the theoretical description of atomic diffraction by laser light.

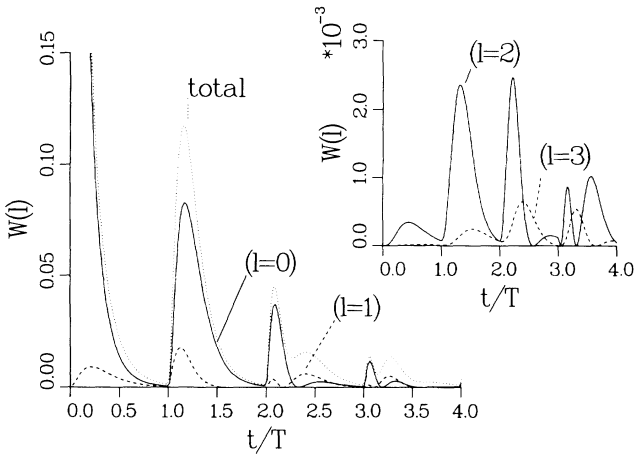

FIG. 2. $W(l) = \mid a_g^{(l)}(t) \mid^2$ as a function of the interaction time $t = L/v_{\text{in}}$ in units of T for $\delta\omega = 0$, $\gamma T = 10.0$, and $e^{2i\pi(\overline{\nu}+\alpha)} = 1.0$. The total initial state probability (total) is also shown.

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[1] A.P. Kazantsev, G.A. Ryabenko, and G.I. Surdutovich, Phys. Rep. **129**, 75 (1985).
[2] For a survey of recent work, see Appl. Phys. B **54**, 321ff (1992), and references therein.
[3] For a treatment of multilevel effects in ladder systems, see M. Lindberg, Appl. Phys. B **54**, 467 (1992).
[4] G. Alber and P. Zoller, Phys. Rep.**199**, 231 (1991).
[5] M.J. Seaton, Rep. Prog. Phys. **46**, 167 (1983).
[6] N.G. van Kampen, *Stochastic Processes in Physics and Chemistry* (North-Holland, Amsterdam, 1981), p. 142ff.
[7] G. Alber, Z. Phys. D **14**, 307 (1989).
[8] Yu.B. Ovchinnikov and V.S. Lethokov, Comments At. Mol. Phys. **27**, 185 (1992).