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Electronic Wavepackets and Deflection of Atoms by Intense Laser Fields

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Abstract – The problem of deflection of Rydberg atoms by intense laser light is approached with the help of a semiclassical path representation, with respect to classical periodic paths of an excited Rydberg electron around the positively charged ionic core. This approach allows the momentum exchange between atom and laser field to be related to elementary laser-assisted collisions between an excited Rydberg electron and the ionic core, and clearly exhibits the "particle" and "wave" aspects of this momentum exchange.

1. INTRODUCTION

The recent development of short and intense laser pulses has aroused much interest in the behavior of atoms and molecules under the influence of intense laser radiation. Depending on the laser intensity, two main dynamical regimes may be distinguished:

(1) For laser intensities that are larger than the atomic unit of intensity $I_0 = 1.4 \times 10^{17}\,\mathrm{W/cm^2}$, the dynamics of atomic or molecular electrons are altered significantly by the laser field, even as far as energetically low-lying bound states are concerned, and the concept of a weakly perturbed "bare atom or molecule" loses its physical significance.

(2) For laser intensities I with $I \ll I_0$, the dynamics of atomic or molecular electrons are only slightly modified by the presence of a laser field. However, a variety of interesting physical phenomena occur in this intensity regime, which cannot be described with the help of perturbative methods. An example of a laser-induced process that takes place in this intensity regime (which will be further discussed) is the excitation of atomic Rydberg states close to a photoionization threshold with laser photons in the optical frequency range [1, 2]. In this type of excitation process, because of the small level spacings of highly excited Rydberg states, even a relatively weak laser field may cause coherent excitation of a large number of Rydberg and continuum states. In such a coherent excitation process, a radial electronic Rydberg wavepacket is prepared, which is well localized in comparison with typical extensions of highly excited Rydberg states. The time evolution of such an electronic wavepacket, under the influence of the Coulomb potential of the positively charged ionic core and the laser field, manifests itself in the time dependence of various atomic transition probabilities.

Recently, theoretical methods for the description of laser-induced excitation processes that involve atomic Rydberg (and possibly continuum) states close to a photoionization threshold have been developed [1 - 3]. They are based on the observation that in the optical frequency regime, and for laser intensities $I \ll I_0$, the atom-laser interaction takes place in a *finite reaction zone* around the atomic nucleus [3]. Typically, this region has an extension of a few Bohr radii, and is therefore small in comparison with the extensions of highly excited Rydberg states. Furthermore, relevant atomic transition amplitudes are expressed in this theory as a sum of probability amplitudes, which are associated with the motion of an excited Rydberg electron along a periodic classical Coulomb orbit with near-zero angular momentum. These *semiclassical path representations* are particularly useful for the description of coherent laser-induced excitation processes where an electronic Rydberg wavepacket is generated. So far, applications of this approach have neglected the effects of the exciting laser field on the atomic center-of-mass motion.

The deflection of atomic beams by laser light has been a problem of atomic physics that has aroused considerable interest (experimentally and theoretically) during the last few years [4, 5]. However, up until now, investigations on this problem have concentrated mainly on two- or few-level systems, or on harmonic-oscillator-like ladder systems [6] concerning the internal dynamics of the atoms. The discussions in this paper concerning the deflection of atoms by intense laser fields is based on recent work on the coherent laser-induced excitation of atomic Rydberg states [1, 2]. It is shown that, with the help of a semiclassical path representation of the relevant state-selective probability amplitudes, the deflection of atoms by laser light can be described in a simple and unified way, that covers cases where many Rydberg states are excited coherently, as well as two-level limit cases where only one Rydberg state is excited significantly during the motion of an atom through a laser beam [7]. In this approach, the relevant transition probability amplitudes are represented as a sum of probability amplitudes that are associated with repeated collisions between the excited Rydberg

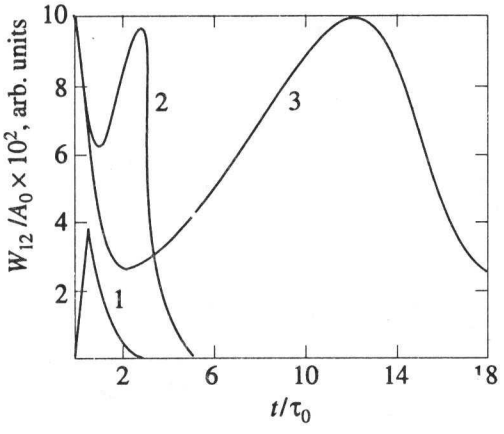

Fig. 8.

$$F(\Omega) = C\exp\{-\frac{(\Delta u - a\hbar\omega - \hbar\Omega)^2}{\hbar^2\omega^2 a(1+2\tilde{n}_F)}\}. \quad (80)$$

For centrosymmetric molecules, calculation of the spectrum is performed in the same way. Figure 7 compares the calculated spectrum (curve 2) of instantaneous luminescence to the experimental data (curve 1) on the spectrum of instantaneous luminescence of the SiF_4 molecules (maximum luminescence is 2.78 eV, half-width is about 1 eV [24]).

Especially interesting is the investigation of optical characteristics of a molecule depending on duration τ_0 of the pulse of IR radiation. In this case (according to (27) and (28)), at a given pulse shape, the mean occupation numbers $\tilde{n}_F(t)$ depend on t. In their turn, optical characteristics of a molecule begin to be dependent on time. Moreover, the time of spectrum recording ΔT must be shorter than the characteristic time of spectrum change. Figure 8 shows the dependence of the probability of the multiquantum transition between electronic levels of different multiplicity on time at different magnitudes of $\gamma\tau_0$ (1 - 0.15, 2 - 2, 3 - 20), and at the energy density 0.6 J/cm^2. As follows from Fig. 8, there should be a certain delay in time of the luminescence peak relative to the peak of radiation of the CO_2 laser [25].

Molecular systems are convenient objects for demonstrating interference effects appearing under bichannel molecular excitation from the ground state (1) into the excited one (2). The first channel is the direct electron transition (e), the second one is the excitation of nuclear subsystem (q) (the excitation of the vibration oscillator with the following nonadiabatic transition to the excited state of a molecule). If the position of the molecule is fixed, for example, on the surface of a solid body, then at a certain incidence angle θ of the linearly polarized radiation of CO_2, laser interference of both channels of excitation is possible. The total amplitude of transition $1 \to 2$ is

$$A_{12}(\theta) = A^e_{12}(\theta) + A^q_{12}(\theta).$$

The amplitude A^e_{12} determines the probability (1), the amplitude A^q_{12} determines the probability of multiquantum transition. In particular, in the model of shifted parabolas, the expression was obtained for the critical angle θ_{cr}, at which the total amplitude (81) is equal to zero.

REFERENCES

1. Franck-Kamenetsky, M.D. and Lukashin, A.V., 1975, *Usp. Fiz. Nauk*, **116**, 193.
2. Kovarsky, V.A., Perelman, N.F., and Averbuch, I.Sh., 1985, *Multiquantum Transitions* (Moscow: Energoatomizdat) (in Russian).
3. Medvedev, E.S. and Osherov, V.I., 1983, *The Theory of Radiationless Transitions in Multiphoton Molecules* (Moscow: Nauka) (in Russian).
4. Fedorov, M.V., 1977, *Zh. Eksp. Teor. Fiz.*, **79**, 134.
5. Sazonov, V.N., 1977, *Teor. Mat. Fiz.*, **31**, 107.
6. Mukamel, S., Oppenheim, J., and Ross, J., 1978, *Phys. Rev. A*, **17**, 1988.
7. Belousov, A.V., Kovarsky, V.A., and Siniavsky, E.P., 1989, *Optical Properties of Molecular Systems in the Low-Frequency Laser Radiation* (Kishinev: Shtiintsa) (in Russian).
8. Zel'dovich, Ya.B., Perelomov, A.M., and Popov, V.S., 1969, *Zh. Eksp. Teor. Fiz.*, **57**, 196.
9. Lax, M., 1968, *Fluctuation and Coherence Phenomena in Classical and Quantum Physics* (New York: Gordon and Breach).
10. Drummond, R.P. and Gardiner, C.W., 1980, *J. Phys. A*, **13**, 2353.
11. Gradshtein, I.S. and Ryzhik, I.M., 1965, *Tables of Integrals, Series and Products* (New York: Academic).
12. Marichev, O.I., 1978, *The Methods of Calculation Integrals from the Special Functions* (Minsk: Nauka i Tekhnika) (in Russian).
13. Bagratashvili, V.N., Kuz'min, M.V., and Letokhov, V.S., 1985, *Chem. Phys.*, **97**, 1362.
14. Fain, V.M., 1972, *Photons and Nonlinear Medias* (Moscow: Sov. Radio) (in Russian).
15. Louisell, W.H., 1964, *Radiation and Noise in Quantum Electronics* (New York: McGraw-Hill).
16. Dushinsky, F., 1937, *Acta Physicochem. (USSR)*, **7**, 551.
17. Malkin, I.A. and Man'ko, V.I., 1979, *Dynamical Symmetries and Coherent States of Quantal Systems* (Moscow: Nauka) (in Russian).
18. Belousov, A.V., Kovarsky, V.A., and Chebotar', V.N., 1992, *Teor. Mat. Fiz.*, **1**, 138.
19. Delone, N.B., Kovarsky, V.A., Masalov, A.V., and Perelman, N.F., 1980, *Usp. Fiz. Nauk*, **131**, 617.
20. Simpson, T.B. and Bloembergen, N., 1983, *Chem. Phys. Lett.*, **100**, 325.
21. Ambartsumyan, R.V., Makarov, B.N., and Puretsky, A.A., 1975, *Pis'ma Zh. Eksp. Teor. Fiz.*, **22**, 96.
22. Letokhov, V.S., 1983, *Nonlinear Selected Photoprocesses in Atoms and Molecules* (Moscow: Nauka) (in Russian).
23. Mirumyants, S.O., Vandukov, E.A., and Kozlov, V.K., 1978, *Izv. Akad. Nauk SSSR*, **42**, 380.
24. Doljikov, B.C., Chekalin, N.V., and Shabanov, A.V., 1978, *Kvantovaya Elektronika*, **5**, 648.
25. Zalesskaya, G.A., 1980, *J. Appl. Spec.*, **32**, 34.

electron and the ionic core. These collisions take place inside the reaction zone. Outside the reaction zone, an excited Rydberg electron moves on a classical periodic Coulomb orbit with near-zero angular momentum. Each collision leads to a momentum exchange between the laser field and the center-of-mass of an excited atom. In cases where many Rydberg states are excited coherently, these collision events are sufficiently separated in time so that the "particle" aspects of the electronic dynamics under the combined influence of the positively charged ionic core and the laser field prevail. In the other limiting case where only one Rydberg state is excited significantly by the laser field, the probability amplitudes of almost all possible laser-assisted electron-ion collisions overlap in time so that the "wave" aspects of the electronic dynamics dominate.

This paper is organized as follows:

In Section 2, a general theoretical framework for the description of laser-induced excitation processes of atomic Rydberg states and for the description of their effect on the atomic center-of-mass motion is developed. A semiclassical path representation is derived for the state-selective probability amplitude of observing an atom after the interaction with the laser field in the initially prepared energetically low lying bound state. In Section 3, this result is specialized to the description of the deflection of fast atoms by a standing-wave laser field. A simple analytical expression is obtained for this state-selective probability amplitude, which is valid in cases where dispersion effects of an excited electronic Rydberg wavepacket may be neglected.

2. PROBLEM AND THEORETICAL FRAMEWORK

The typical layout of an atomic diffraction experiment is shown in Fig. 1. A beam of atoms with well-defined initial momenta $\mathbf{P}_{in}$, which are prepared initially in an energetically low lying bound state $|g\rangle$, crosses a laser beam with electric field strength $\mathbf{E}(\mathbf{x}, t) = \mathbf{e}\varepsilon(\mathbf{x})e^{-i\omega t} + \text{c.c.}$ ($\mathbf{e}$ is the polarization and ω is the frequency of the laser field). During its flight through the laser field the atom becomes excited. By this excitation process, momentum is transferred from the laser field to the atomic center of mass, and the atom may leave the interaction region in different directions. In the following, I am interested in cases where a large number of Rydberg states $|n\rangle$ might be excited resonantly from the initial state $|g\rangle$ during the motion of the atom through the laser field. For simplicity this paper considers only alkali atoms with one valence electron and a (rigid) spherically symmetric core. I assume that their energies are determined by an energy-independent quantum defect α. Furthermore, in the following I ignore ionization from the excited Rydberg states to continuum states well above threshold. This is valid for sufficiently small interaction times (see also the remark at the end of Section 3). These effects may be taken into account with the help of additional continuum channels [1].

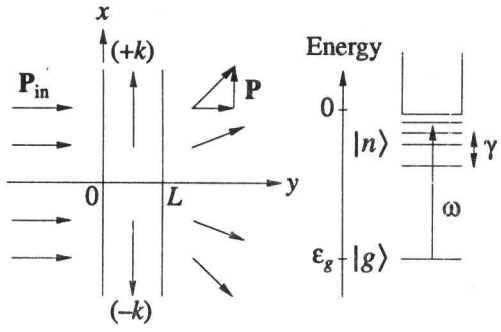

Fig. 1. *Schematic representation of the atomic diffraction experiment and the corresponding laser-induced excitation process ($\gamma = \Gamma/\varepsilon_0^2$).*

Disregarding the direct influence of the laser field on the atomic center-of-mass motion, the Hamiltonian, which describes the dynamics of an atom as it moves through the laser field, is given by

$$\hat{H} = \frac{\hat{\mathbf{P}}^2}{2M} + \hat{H}_A - \hat{\vec{\mu}}\,\mathbf{E}(\mathbf{x}, t)\,, \tag{1}$$

with the atomic Hamiltonian $\hat{H}_A$, the atomic mass M, the atomic dipole operator $\hat{\vec{\mu}}$, and the momentum operator $\hat{\mathbf{P}}$, which is associated with the atomic center-of-mass coordinates $\mathbf{x}$. In the following, I use Hartree atomic units. The state of the atom at time t is closely approximated as

$$\langle \mathbf{x}|\psi\rangle_t = a_g^{(t)}(\mathbf{x})\,|g\rangle + \sum_n a_n^{(t)}(\mathbf{x})\,|n\rangle, \tag{2}$$

with the sum over n indicating summation and integration over all bound and continuum states. In the dipole and rotating-wave approximation, we obtain from the time-dependent Schrödinger equation the equations

$$\left(z - \frac{\hat{\mathbf{P}}^2}{2M} - e_g\right) a_g^{(z)}(\mathbf{x}) + \sum_n \langle g|\,\vec{\mu}\,\mathbf{e}^*\varepsilon^*(\mathbf{x})\,|n\rangle \times a_n^{(z)}(\mathbf{x}) = i a_g^{(t=0)}(\mathbf{x}), \tag{3}$$

$$\left(z + \omega - \frac{\hat{\mathbf{P}}^2}{2M} - e_n\right) a_n^{(z)}(\mathbf{x}) + \langle n|\,\vec{\mu}\,\mathbf{e}\varepsilon^*(\mathbf{x})\,|g\rangle \times a_g^{(z)}(\mathbf{x}) = i a_n^{(t=0)}(\mathbf{x}) \tag{4}$$

for the Laplace transforms $a_j^{(z)}(\mathbf{x})$ $(j = g, n)$ of the atomic probability amplitudes. They are defined by

$$a_j^{(z)}(\mathbf{x}) = \int_0^\infty dt e^{i(z+i0)t} a_j^{(t)}(\mathbf{x}) \tag{5}$$

and the inverse relation

$$a_j^{(t)}(\mathbf{x}) = \frac{1}{2\pi}\int_{-\infty+i0}^{\infty+i0} dz e^{-izt} a_j^{(z)}(\mathbf{x}), \qquad (6)$$

with $j = g, n$. The amplitudes $a_g^{(t=0)}(\mathbf{x})$ and $a_n^{(t=0)}(\mathbf{x})$ describe the initial condition of the atom as it enters the laser field at time $t = 0$. Inserting (4) into (3), and assuming that the atom is initially prepared in state $|g\rangle$, we obtain the equation

$$\left[z - \frac{\hat{\mathbf{P}}^2}{2M} - e_g - \hat{\Sigma}(z)\right] a_g^{(z)}(\mathbf{x}) = i a_g^{(t=0)}(\mathbf{x}) \qquad (7)$$

for the Laplace transform of the initial state probability amplitude. The quantity

$$\hat{\Sigma}(z) = \langle g|\vec{\mu}\,\mathbf{e}^*\varepsilon^*(\mathbf{x})\left[z + \omega - \frac{\hat{\mathbf{P}}^2}{2M} - \hat{H}_A\right]^{-1} \times \vec{\mu}\mathbf{e}\varepsilon^*(\mathbf{x})\,|g\rangle \qquad (8)$$

is the self-energy operator of the initial state $|g\rangle$. In general, $\hat{\Sigma}(z)$ is a complicated nonlocal operator. More particularly, in cases where many Rydberg states are excited coherently by the laser field, a direct physical interpretation of (7) and its inverse Laplace transform is difficult. A great deal of physical insight into the elementary processes that lead to the deflection of atoms may be obtained by deriving a semiclassical path representation for (7). Thereby, the transition probability amplitude is expressed as a sum of probability amplitudes associated with repeated returns of an excited Rydberg electron to the reaction zone where the atom-laser interaction is localized. For the problem considered here, such a semiclassical path representation may be derived with the help of the Poisson sum formula and the results of the Quantum Defect Theory [8].

As outlined in the Appendix, from (7) we obtain the semiclassical path representation

$$a_g^{(z)}(\mathbf{x}) = \left[\hat{U}_0(z) - i\Gamma\hat{U}_0(z)\,\varepsilon^*(\mathbf{x})e^{2i\pi(\hat{\nu}+\alpha)} \times \sum_{N=1}^{\infty}\left(\hat{\tilde{\chi}}(z)\,e^{2i\pi\hat{\nu}}\right)^{N-1}\varepsilon(\mathbf{x})\hat{U}_0(z)\right] i a_g^{(t=0)}(\mathbf{x}), \qquad (9)$$

with

$$\hat{U}_0(z) = \left[z - \frac{\hat{\mathbf{P}}^2}{2M} - e_g - (\delta\omega - i\Gamma/2)\,|\varepsilon(\mathbf{x})|^2\right]^{-1}. \qquad (10)$$

The quantum defect of the Rydberg states is denoted by α and $\hat{\nu} = \int d^3P\,|\mathbf{P}\rangle\langle\mathbf{P}|\,[-2(z+\omega-\mathbf{P}^2/2M)]^{-1/2}$. The physical interpretation of the terms on the right-hand side of (9) is straightforward.

Apart from the initial condition, the first term on the right-hand side of (9) is determined only by the resolvent operator $\hat{U}_0(z)$. It describes the dynamics of the atomic center of mass in the optical potential $(\delta\omega - i\Gamma/2)|\varepsilon(\mathbf{x})|^2$, which originates from the depletion of the initially prepared atomic state $|g\rangle$ by the laser field. The quantities $\delta\omega|\varepsilon(\mathbf{x})|^2$ and $\Gamma|\varepsilon(\mathbf{x})|^2$ are the quadratic Stark shift (due to virtual transitions between $|g\rangle$ and all other nonresonant excited states) and the ionization rate, respectively. According to Fermi' golden rule, the term $\Gamma|\varepsilon(\mathbf{x})|^2$ describes ionization from the initial state $|g\rangle$ to continuum states close to the threshold. The quantities $\delta\omega$ and Γ are defined in Eq. (A.4) of the Appendix. Both quantities characterize the atom-laser interaction, which is localized in a finite reaction zone around the atomic nucleus. As this reaction zone is small in comparison with the extension of highly excited Rydberg states, they are approximately independent of z and $\mathbf{P}^2/2M$.

The remaining terms on the right-hand side of (9) are probability amplitudes, which are associated with repeated returns of the excited Rydberg electron to the reaction zone. The N-th term describes the contribution of the N-th return. The quantity $2\pi\langle\mathbf{P}|\hat{\nu}|\mathbf{P}\rangle$ is the classical action of a periodic Coulomb orbit with near-zero angular momentum and energy $e = z + \omega - \mathbf{P}^2/2M$. With each return to the reaction zone, an excited Rydberg electron may either be deexcited to the initial state $|g\rangle$ by stimulated emission of a laser photon, or may be scattered by the ionic core in the presence of the laser field. This laser-assisted electron-ion scattering process is described by the scattering operator

$$\hat{\tilde{\chi}}(z) = \chi_c\,[1 - i\Gamma\varepsilon(\mathbf{x})\hat{U}_0(z)\,\varepsilon^*(\mathbf{x})]\,. \qquad (11)$$

The scattering matrix element $\chi_c = e^{2i\pi\alpha}$ describes scattering of the Rydberg electron by the spherically symmetric ionic core [8].

Inserting the semiclassical path representation of (9) into (6), we obtain the probability amplitudes $a_g^{(t)}(\mathbf{x})$ and $a_n^{(t)}(\mathbf{x})$, and, thus, a complete description of the laser-induced excitation process. Although the semiclassical path representation allows one to split the state vector of the atom $\langle\mathbf{x}|\psi\rangle_t$ into contributions that originate from repeated returns of the excited Rydberg electron to the reaction zone, it is still difficult to come up with an explicit evaluation under general conditions. In the next section, we will study a model problem where (9) can be simplified considerably and a simple analytical expression may be obtained for the state-selective probability amplitude $a_g^{(t)}(\mathbf{x})$.

3. DEFLECTION OF ATOMS BY A STANDING-WAVE LASER FIELD

For the special case of fast atoms, a simple analytical expression is obtained for the state-selective transition probability amplitude $a_g^{(t)}(\mathbf{x})$, which clearly exhibits the particle (wavepacket) and wave (two-level) aspects

of the momentum exchange between the laser field and the center of mass of an excited atom.

For this purpose, assume that the laser field is approximated as

$$\varepsilon(\mathbf{x}) = \Theta(y)\Theta(L-y)\varepsilon(x), \tag{12}$$

with $\varepsilon(x) = \varepsilon_0 \sin kx$ and the wavenumber $k = \omega/c$ (compare with Fig. 1). Furthermore, assume that the atoms enter the laser field with well-defined momenta $P_y = P_{in}$ and $P_x = P_z = 0$. The initial momentum P_{in} is assumed to be so large that the atomic motion along the y direction is not influenced significantly by the optical potential $(\delta\omega - i\Gamma/2)\,|\varepsilon(x)|^2$, i.e.,

$$\frac{P_{in}^2}{2M} \gg \left|(\delta\omega - i\Gamma/2)\,|\varepsilon_0|^2\right|, \tag{13}$$

and that during the flight of an atom through the laser field the atomic motion in the x-direction can also be neglected, i.e.,

$$\frac{P_x}{P_{in}} L \ll \lambda = \frac{2\pi}{k}. \tag{14}$$

Under present conditions in (9), the operator of kinetic energy $\hat{\mathbf{P}}^2/2M$ may be replaced by $P_{in}^2/2M$. These assumptions are equivalent to the eikonal approximation.

The inversion of the Laplace transform of (6) is facilitated if the energy range $(e_g + \omega - \Gamma|\varepsilon_0|^2/2,\ e_g + \omega + \Gamma|\varepsilon_0|^2/2)$, which is excited significantly by the laser field, is located well below the photoionization threshold and if the ionization rate $\Gamma|\varepsilon_0|^2$ is not too large, i.e.,

$$\pi N \left| \frac{d^2\nu}{dz^2} (\delta\omega - i\Gamma/2)^2 |\varepsilon_0|^4 \right|_{z = e_g + P_{in}^2/2M} \ll 1. \tag{15}$$

This implies that the dispersion of an electronic Rydberg wavepacket generated during the laser-induced excitation process is neglected. Thus, the classical action of a periodic Coulomb orbit may be linearized with respect to energy,

$$2\pi\hat{\nu} \approx 2\pi\bar{\nu} + T\left(z - e_g - \frac{P_{in}^2}{2M}\right), \tag{16}$$

with $\bar{\nu} = [-2(e_g + \omega)]^{-1/2}$ and the mean classical orbit time of the excited Rydberg states $T = 2\pi\bar{\nu}^3$. Using the expansion

$$[\hat{\chi}(z)]^N = \chi_c^N \sum_{r=0}^{N} \binom{N}{r} [-i\Gamma\varepsilon(\mathbf{x})\hat{U}_0(z)\,\varepsilon^*(\mathbf{x})]^r, \tag{17}$$

we thus find from (9) and (6), within the approximations of (13) - (15) and the residue theorem, the semiclassical path representation

$$\begin{aligned} a_g^{(t)}(P) = e^{-i\left(e_g + \frac{P_{in}^2}{2M}\right)t} \langle P| \{ & e^{-i(\delta\omega - i\Gamma/2)|\varepsilon(x)|^2 t} \\ & + \sum_{N=1}^{\infty} \Theta(t - NT) e^{2\pi i(\bar{\nu} + \alpha)N} e^{-i(\delta\omega - i\Gamma/2)|\varepsilon(x)|^2 (t - NT)} \\ & \times \sum_{r=0}^{N-1} \binom{N-1}{r} \frac{[-\Gamma|\varepsilon(x)|^2 (t - NT)]^{r+1}}{(r+1)!} \} \, |P_x = 0\rangle. \end{aligned} \tag{18}$$

This is the main result of this section. An equivalent, more explicit form is given in (20). It expresses the state-selective probability amplitude $a_g^{(t)}(P)$ of finding an atom at time $t = LM/P_{in}$ after the interaction with the laser field in the initial state $|g\rangle$, with momentum P along the x-direction, as a sum of probability amplitudes associated with repeated collisions between an excited Rydberg electron and the laser field. These collisions take place inside the finite reaction zone around the atomic nucleus.

The first term on the right-hand side of (18) describes the momentum transfer between the center of mass of an excited atom and the laser field, which is due to depletion of the initially prepared state $|g\rangle$. For interaction times $t < T$ it is the only contribution to $a_g^{(t)}(P)$. The standing wave of (12) implies that, in momentum space, the matrix representation of the optical potential is proportional to the transfer matrix of an unbounded symmetrical random walk [9]. Therefore, this first term describes a random walk in momentum space with the complex-valued transition rate $(\Gamma/2 + i\delta\omega)|\varepsilon_0|^2/4$. Consequently, the state-selective probability amplitude of (18) can only have nonzero amplitudes with momenta $P = \pm 2kl$ $(l = 0, 1, 2, \ldots)$. This reflects the fact that for the observation of an atom in the initial state $|g\rangle$ after the interaction with the laser field, an equal number of photon absorptions and stimulated emissions is required, each of which may transfer a momentum of magnitude $\pm k$ along the x-direction to the center of mass of an excited atom.

The remaining terms on the right-hand side of (18) are associated with repeated returns of the excited Rydberg electron to the finite reaction zone around the atomic nucleus. According to (18), outside the reaction zone the excited Rydberg electron moves along one of the classical periodic Coulomb orbits of near-zero angular momentum, whose classical action is given by $S = 2\pi\bar{\nu}$. The additional phase contribution of magnitude $2\pi\alpha$ originates from elastic scattering of the excited Rydberg electron by the spherically symmetric ionic core [8]. The integer N counts the number of returns of the excited Rydberg electron to the reaction zone. The unit step function $\Theta(t - NT)$ ensures that the N-th term only contributes for interaction times $t > NT$. With each crossing of the reaction zone, the laser-induced coupling between initial state $|g\rangle$ and excited states $|n\rangle$ is turned on and off again. Thus, by stimulated emission from the excited Rydberg states and sub-

sequent photon absorption, two laser photons are exchanged during each of these laser-assisted electron-ion collisions. Consequently, the momentum of the center of mass of an excited atom is changed by an amount $\Delta P = 0$ or $\Delta P = \pm 2k$ along the x-direction because these photons can have the same or opposite momenta k. The (N, r)-th term in (18) is the probability amplitude of the N-th return of the excited Rydberg electron to the reaction zone, with a total number of r laser-assisted electron-ion scatterings during intermediate returns to the reaction zone. With each of these scatterings, the excited Rydberg electron experiences a time delay that is described by the factor

$$[-\Gamma|\varepsilon(x)|^2(t-NT)]^r \; e^{-i(\delta\omega - i\Gamma/2)|\varepsilon(x)|^2(t-NT)}. \qquad (19)$$

The binomial coefficient $\binom{N-1}{r}$ is equal to the number of ways that r scattering events of this type can be selected from a total number of $(N-1)$ intermediate returns of an excited Rydberg electron to the reaction zone.

Within the limits of the approximations of (13) - (15), equation (18) gives a unified description of the momentum exchange between the atomic center of mass and the laser field, which is valid in the two-level limit where only one Rydberg state is excited significantly, i.e., where $[\Gamma|\varepsilon_0|^2]^{-1} \gg T$, as well as in the opposite limit where many Rydberg states are excited coherently, i.e., where $[\Gamma|\varepsilon_0|^2]^{-1} < T$, and an electronic wavepacket is generated [1, 2]. The two-level limit describes the wave aspects of the problem because, in this case, state-selective probability amplitudes associated with almost all returns of an excited Rydberg electron to the reaction zone overlap in time and also interfere. In the opposite limit where an electronic Rydberg wavepacket is generated, the particle aspect of the momentum transfer between atomic center-of-mass motion and laser field prevails because, at least for sufficiently small interaction times, probability amplitudes associated with repeated returns of the excited electron to the reaction zone are sufficiently separated in time.

Finally, from (18) a more explicit expression may be derived for the state-selective probability amplitude $a_g^{(t)}(l)$ of observing the atom after the interaction with the laser field in the initial state $|g\rangle$ with momentum $P = 2lk$ along the x-direction, namely,

$$a_g^{(t)}(l) = e^{-i\left(\varepsilon_g + \frac{P_{in}^2}{2M}\right)t} \{ I_l(t) + \sum_{N=1}^{\infty} \Theta(t-NT) e^{2i\pi(\bar{\nu}+\alpha)N} \times \sum_{r=0}^{N-1} \binom{N-1}{r} \frac{[-\Gamma|\varepsilon_0|^2(t-NT)]^{r+1}}{(r+1)!\,4^{-(r+1)}} \times \sum_{\mu=0}^{2(r+1)} \binom{2(r+1)}{\mu} (-1)^{\mu} I_{l+r+1-\mu}(t-NT) \}, \qquad (20)$$

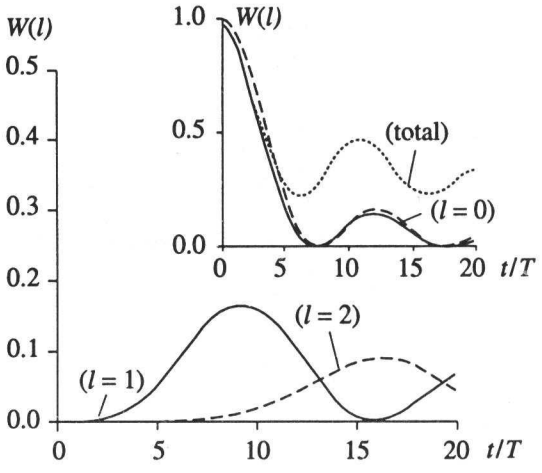

Fig. 2. *Dependence of* $|a_g^{(t)}(l)|^2$ *and* $\sum_{l=-\infty}^{\infty}|a_g^{(t)}(l)|^2$ *(total) as evaluated from (20) on the interaction time* $t = LM/P_{in}$ *(in units of the mean classical orbit time T) for* $\Gamma|\varepsilon_0|^2 T = 0.1$ *and resonant excitation, i.e.,* $e^{2i\pi(\bar{\nu}+\alpha)} = 1$.

where

$$I_l(t) = e^{-i(\delta\omega - i\frac{\Gamma}{2})|\varepsilon_0|^2\frac{t}{2}} e^{i\frac{\pi l}{2}} J_l\left[(\delta\omega - i\frac{\Gamma}{2})|\varepsilon_0|^2\frac{t}{2}\right],$$

and $J_l[x]$ [10] is the Bessel function of integer order.

Figure 2 shows the probability of detecting an atom after the interaction with the laser field in its initially prepared state with momentum $P_x = 2lk$ as a function of the interaction time $t = LM/P_{in}$. In this case, during the flight of an atom through the laser field, essentially only one Rydberg state is resonantly excited, because $\Gamma|\varepsilon_0|^2 T < 1$. Therefore, Eq. (20) is expected to reduce to the corresponding result of a resonantly excited two-level system [4], i.e.,

$$a_g^{(t)}(l) = J_{2l}(\Omega t), \qquad (21)$$

with the Rabi frequency $\Omega = \sqrt{\Gamma|\varepsilon_0|^2/T}$. In the inset of Fig. 2, the initial state probability for $l = 0$ as evaluated from (20) (full line) is compared with the corresponding result of the two-level limit of (21) (dashed line). Because in this case the initial-state probability amplitudes of many laser-assisted collisions between the electron and the ionic core overlap in time, the wave aspects of the resulting momentum transfer between laser field and the center of mass of the excited atom prevail. In particular, as is apparent in Fig. 2, this implies that $|a_g^{(t)}(l)|^2$ changes slowly on the time scale determined by the time interval T between two such collisions.

Figure 3 shows a case where the initial atomic state $|g\rangle$ is depleted on a time scale brief in comparison with the mean classical orbit time of the excited Rydberg states, i.e., $[\Gamma|\varepsilon_0|^2]^{-1} < T$. Thus, a radial electronic Rydberg wavepacket is generated by the laser-induced excitation process, and the particle aspect of the

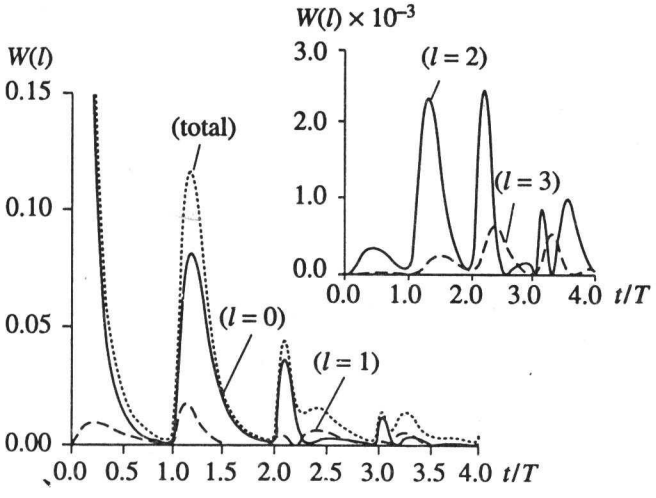

Fig. 3. *The same quantities as in Fig. 2 but with parameter* $G|\varepsilon_0|^2T = 10.0$.

momentum exchange between the center-of-mass motion of an excited atom and the laser field becomes apparent. This implies that a significant momentum transfer takes place only at interaction times where the excited Rydberg electron is inside the reaction zone close to the atomic nucleus.

Finally, I want to point out once more that in the model considered here, ionization from the excited Rydberg states to continuum states well above threshold has been neglected. This is valid as long as relevant interaction times t are sufficiently small in the sense that $\Gamma|\varepsilon_0|^2 t/T \ll 1$.

CONCLUSION

Compared with conventional approaches, which are based on dressed-state analysis [6], the semiclassical path representation discussed in this paper offers the conceptual advantage of relating relevant quantum mechanical transition probability amplitudes directly to elementary laser-assisted electron-ion collisions. Each of these collisions leads to an exchange of momentum between the laser field and the center of mass of an excited atom. This allows us to describe the particle (wavepacket) aspects and the wave (two- or few-level) aspects of this momentum exchange in a unified way. Furthermore, this approach offers the additional advantage that electron-correlation effects (couplings between Rydberg series and continua) [8] and effects of additional static external fields can be taken into account in a natural way [11].

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APPENDIX

In this appendix I outline the derivation of (9).

The main problem is the derivation of a semiclassical path representation for the self-energy operator of (8), i.e.,

$$\hat{\Sigma}(z) = \int d^3P\, \varepsilon^*(\mathbf{x})\, |\mathbf{P}\rangle \sum_n \frac{|\langle n|\, \hat{\mu}\mathbf{e}\, |g\rangle|^2 \langle \mathbf{P}|\, \varepsilon(\mathbf{x})}{z + \omega - \mathbf{P}^2/2M - e_n}, \quad \text{(A.1)}$$

with the sum over states $|n\rangle$ indicating a sum over all bound and continuum states. According to standard results of the Quantum Defect Theory [8] close to the threshold, the energy dependence of the dipole matrix elements in (A.1) is given by

$$|\langle n|\, \hat{\mu}\mathbf{e}\, |g\rangle|^2 = |\langle e|\, \hat{\mu}\mathbf{e}\, |g\rangle|^2 \left(\frac{de_n}{dn}\right), \quad \text{(A.2)}$$

with $e_n = -1/[2(n - \alpha)]^2$ and the energy-normalized dipole matrix element $\langle e|\, \hat{\mu}\mathbf{e}\, |g\rangle$, which is independent of energy, approximately. Using this approximation and the Poisson sum formula for performing the summation over all Rydberg states, we obtain

$$\hat{\Sigma}(z) = \int d^3P\, \varepsilon^*(\mathbf{x})\, |\mathbf{P}\rangle \times \left[\delta\omega - i\Gamma/2 - i\Gamma \sum_{N=1}^{\infty} e^{2i\pi(\nu+\alpha)N}\right] \langle \mathbf{P}|\, \varepsilon(\mathbf{x}), \quad \text{(A.3)}$$

with $-1/2\nu^2 = z + \omega - \dfrac{\mathbf{P}^2}{2M}$. This is the required semiclassical path representation for the self-energy operator. The N-th term in the sum of (A.3) may be interpreted as the contribution of the N-th return of a Rydberg electron of energy $e = -1/2\nu^2$ to the core region. The quadratic Stark shift $\delta\omega|\, \varepsilon(\mathbf{x})\,|^2$, which is due to virtual transitions between the initial state $|g\rangle$, and all other nonresonant atomic states and the ionization rate $\Gamma|\, \varepsilon(\mathbf{x})\,|^2$ are determined by the relation

$$\delta\omega - i\pi\Gamma = \int_{-\infty}^{\infty} \frac{de\, |\langle e|\, \hat{\mu}\mathbf{e}\, |g\rangle|^2}{\left[z + i0 + \omega - \dfrac{\mathbf{P}^2}{2M} - e\right]}. \quad \text{(A.4)}$$

Substituting (A.3) into (7) we obtain (9).

REFERENCES

1. Alber, G. and Zoller, P., 1988, *Phys. Rev. A*, **37**, 377.
2. Alber, G. and Zoller, P., 1991, *Phys. Rep.*, **199**, 231.
3. Giusti, A. and Zoller, P., 1987, *Phys. Rev. A*, **36**, 5178.
4. Kazantsev, A.P., Ryabenko, G.A., and Surdutovich, G.I., 1985, *Phys. Rep.*, **129**, 75.
5. For a survey of recent work see, 1992, *Appl. Phys. B*, **54**, 321ff and references therein.
6. Lindberg, M., 1992, *Appl. Phys. B*, **54**, 467.
7. Alber, G., 1992, *Phys. Rev. Lett.*, **69**, 3045.
8. Seaton, M.J., 1983, *Rep. Prog. Phys.*, **46**, 167.
9. van Kampen, N.G., 1981, *Stochastic Processes in Physics and Chemistry*, (North Holland, Amsterdam), 142ff.
10. *Handbook of Mathematical Functions*, M. Abramowitz and I.A. Stegun, Eds., 1964, National Bureau of Standarts (Washington, DC: USGPO).
11. Alber, G., 1989, *Z. Phys. D*, **14**, 307.