Destruction of quantum coherence and stochastic ionization of Rydberg electrons by fluctuating laser fields

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Abstract: It is shown that diffusion and stochastic ionization of an optically excited Rydberg electron are generic long time phenomena which are consequences of the destruction of quantum coherence by laser fluctuations. Quantitatively these novel fluctuation-induced phenomena are characterized by non-exponential time evolutions whose power law dependences can be determined analytically. It is demonstrated that the competition between stochastic ionization and autoionization may lead to interesting new effects.

OCIS codes: (020.5780) Rydberg states; (020.1670) Coherent optical effects; (020.4180) Multiphoton processes

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The rapid development of new experimental techniques particularly in the fields of atomic, molecular and optical physics have made possible the controlled preparation and observation of wave packets of elementary constituents of matter in real time. Within the past decade these advancements have led to numerous investigations[1] on

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electronic wave packets and atomic (center-of-mass) wave packets in atoms, molecules, clusters and nanostructures. These wave packets are quantum states which are situated on the border between the microscopic and macroscopic regime and the understanding of their dynamics is essential for our conception of quantum mechanics and of its connection with classical mechanics. So far studies on the dynamics of wave packets have concentrated mainly on semiclassical aspects which may be attributed to the smallness of the relevant De Broglie wave lengths. Thereby quantum aspects still manifest themselves in interferences between probability amplitudes which are associated with those (families of) classical trajectories along which probability amplitudes are propagated. However, for a comprehensive understanding of the emergence of classical behaviour in wave packet dynamics it is also essential to investigate the dynamical consequences of destruction of coherence which may be caused by stochastic external forces.

A paradigm in which fundamental consequences of the destruction of quantum coherence on the dynamics of wave packets can be investigated in detail is the resonant, optical excitation of Rydberg electrons by fluctuating laser fields. Due to the inherent stochastic nature of laser light the resonant interaction of atoms and molecules with fluctuating laser fields is one of the central problems of laser spectroscopy. In particular, for interaction times which are large in comparison with the inverse bandwidth b laser fluctuations cannot be suppressed and are unavoidable. Much attention has already been paid to the resonant excitation of isolated, bound atomic or molecular energy eigenstates by intense fluctuating laser fields[2–4]. However, so far scarcely anything is known about cases in which the number of significantly excited energy eigenstates is so large that the nonperturbative interaction of an atom or molecule with a fluctuating laser field exhibits signatures of wave packet dynamics. Particularly for sufficiently long interaction times a variety of novel fluctuation-induced dynamical phenomena are expected to occur which reflect generic features of the destruction of quantum coherence in wave packet dynamics.

Let us first of all briefly summarize basic physical aspects concerning the interaction of Rydberg electrons with laser fields. This process is governed by two characteristic features[1], namely
(1) the localization of the electron-laser interaction in a region which typically extends only a few Bohr radii around the atomic nucleus[5] and
(2) the large extension of the classically accessible region of space in which the dynamics of the Rydberg electron is dominated by the $(1/r)$ Coulomb potential of the positively charged ionic core[6,7].
Property (1) is valid for electromagnetic fields in the optical frequency regime as long as the intensities I are small on the atomic scale, i.e. $I \ll I_0 = 1.40 \times 10^{17} \mathrm{Wcm}^{-2}$. It implies that the dynamics of a Rydberg electron can be affected by a laser field and its fluctuations only as long as it is sufficiently close to the atomic nucleus. In contrast, the dynamical behaviour of a Rydberg electron under the influence of microwave radiation[8,9] is completely different. Due to the smallness of typical excitation frequencies in this latter case the influence of the electromagnetic field is not restricted to a small region around the atomic nucleus so that in this frequency regime photons can be absorbed by a Rydberg electron essentially anywhere within its classically accessible region of space. Property (2) implies that apart from the dynamics inside the core region Rydberg electrons exhibit universal behaviour which can be observed in atoms, molecules or even more complex systems such as clusters. Furthermore, as a consequence of these two main features Rydberg systems can be described theoretically in a systematic and satisfactory way by combining quantum defect theory (QDT)[6,7] with semiclassical methods[1].

As an example for the great variety of novel fluctuation-induced phenomena let us consider the process of one-photon excitation of atomic Rydberg and continuum states close to an ionization threshold by a fluctuating cw-laser field. In the dipole and

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adiabatic approximation (rotating wave approximation) this optical excitation process is described by a Hamiltonian of the form (using atomic units with $\mathrm{e} = \hbar = \mathrm{m}_e = 1$)

$$H = \epsilon_g |g\rangle\langle g| + \sum_{n,i} \epsilon_{n,i} |n,i\rangle\langle n,i| - \sum_{n,i} (|n,i\rangle\langle g|\langle n,i|\mathbf{d}|g\rangle \cdot \mathbf{E}(t) e^{-i\omega t} + \mathrm{h.c.}). \qquad (1)$$

Thereby $\mathbf{d}$ denotes the atomic dipole operator. The initially prepared energetically low lying bound state with energy ϵ_g is denoted $|g\rangle$ and $|n,i\rangle$ are the excited Rydberg and continuum states in channel i with energies $\epsilon_{n,i}$, respectively. The excited Rydberg and continuum states can be described with the help of QDT[6]. In the simplest case of a one-channel approximation the optically excited channels are character ized by complex quantum defects $\mu_i = \alpha_i + i\beta_i$ whose real parts α_i determine the energy eigenstates, i.e. $\epsilon_{n,i} = -1/[2(n-\alpha_i)]^{-2}$, and whose imaginary parts β_i describe depletion of the Rydberg states due to autoionization or due to laser-induced ionization into continuum states well above threshold[1]. Fluctuations of an ideal single mode laser which is operated well above the laser threshold can be described in a realistic way within the framework of the phase diffusion model (PDM)[10]. According to this model the laser field is characterized by a classical stochastic electric field whose amplitude is wellstabilized and whose phase diffuses according to a real-valued Wiener process. Thus the slowly varying envelope of the electric field of Eq. (1) is given by $\mathbf{E}(t) = \mathbf{E}_0 e^{i\Phi(t)}$. The real-valued Wiener process for the phase $\Phi(t)$ is defined by the stochastic differential equations $M d\Phi(t) = 0$ and $[d\Phi(t)]^2 = 2b dt$ (M denotes averaging over the statistical ensemble). The PDM implies a Lorentzian spectrum of the laser field which is centered around the optical frequency ω with bandwidth $b \ll \omega$.

In order to obtain a quantitative understanding of characteristic phenomena originating from the destruction of quantum coherence by laser fluctuations one has to solve the time dependent Schrödinger equation with the stochastic Hamiltonian H of Eq. (1). Thereby a main problem is the simultaneous treatment of the laser fluctuations and the threshold phenomena which originate from the infinitely many bound Rydberg states converging to the photoionization threshold. It has been demonstrated recently[11] that the resulting difficulties can be circumvented successfully. It is not only possible to obtain numerical solutions which are highly accurate even in the long time limit but it is also possible to derive analytical results for the nontrivial long time dynamics. This theoretical approach relies on the fact that within the framework of the PDM a master equation can be derived for the atomic density operator which is averaged over the laser fluctuations and which describes relevant parts of the dynamics of the optical excitation process[12]. With the help of QDT Fourier representations can be developed for the general solution of this master equation from which the long time asymptotics can be determined even analytically. A detailed presentation of this theoretical approach has been presented elsewhere[11]. In the following we primarily focus on a physical discussion of characteristic novel fluctuation-induced effects which are predicted by this general theoretical treatment.

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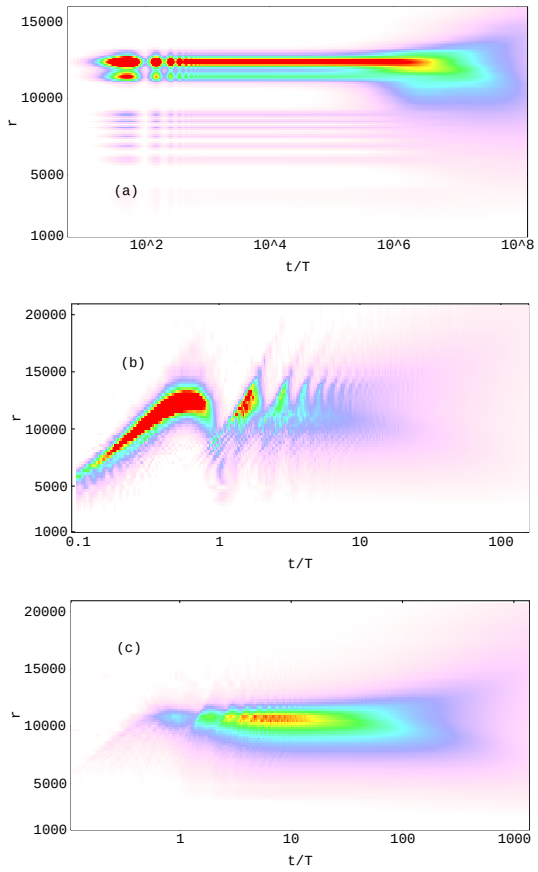

Fig.1: Radial probability distributions of the excited Rydberg electron as a function of interaction time t in units of the mean classical orbit time T (r denotes the radial distance of the Rydberg electron from the nucleus in units of the Bohr radius); $\gamma T = 0.1$, $bT = 0.01$ (a), $\gamma T = 10.0$, $bT = 10.0$ (b), $\gamma T = 0.5$, $bT = 15.0$ (c). (dark red...high probability, light blue...low probability)

Basic qualitative aspects of the time evolution of an excited Rydberg electron are shown in the contour plots of Figs.1a-c. The depicted radial probability distributions refer to one-photon excitation of a hydrogen atom by linearly polarized laser light with $|g\rangle = |2s\rangle$. The principal quantum number of the mean excited state is $\overline{n} = (-2\overline{\epsilon})^{-1/2} = 80$ with the mean excited energy $\overline{\epsilon} = \epsilon_g + \omega$. For the laser intensities and interaction times considered it can be shown that laser-induced ionization from the excited Rydberg states is negligible[11]. In Fig.1a both the laser bandwidth b and the field induced depletion rate of the initial state γ are small in comparison with the mean level spacing of the excited Rydberg state. Thus it is expected that this excitation process is described well within the framework of a two-level approximation. Indeed, the early stages of this optical excitation process are dominated by Rabi oscillations of the electron between initial and excited state. These Rabi oscillations are damped by the laser fluctuations and an equilibrium distribution is approached for interaction times t of the order of $t \geq 1/b$. This behaviour is also apparent from Fig.2a where the time evolution of the corresponding initial state probability $P_g(t)$ is depicted.

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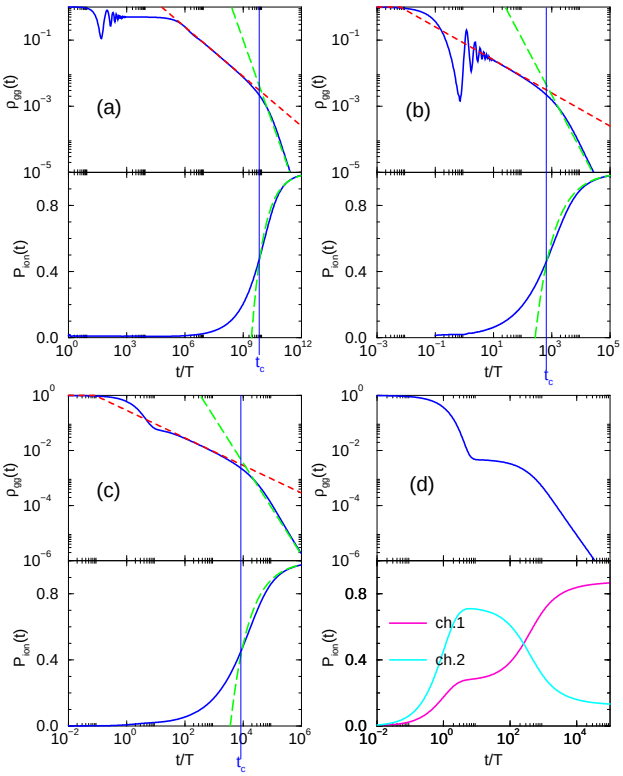

Fig.2: Initial state probability $P_g(t)$ and ionization probability $P_{ion}(t)$ as a function of interaction time t in units of the mean classical orbit time T; parameters as in Fig.1a (a), parameters as in Fig.1b (b), parameters as in Fig.1c (c). In Fig.2d $P_g(t)$ and the ionization probabilites $P_{ion-ch.1}(t)$ and $P_{ion-ch.2}(t)$ of channels 1 and 2 are shown for one-photon excitation of autoionizing Rydberg states with $\overline{n} = \alpha_1 + (-2\overline{\epsilon})^{-1/2} = 80$, $\alpha_1 = 0.1$, $\gamma T = 1.0$, $bT = 300.0$, $\Gamma_n = 2\tau(n-\alpha_1)^{-3}/\pi$ and $\tau = 10^{-5}$a.u..

This equilibrium state of the excited atom is characterized by equal probabilities of the initial and final state and by vanishingly small coherences. However, this equilibrium is only metastable. At interaction times t of the order of $t \geq t_1 \approx 5 \times 10^5 T$ ($T = 2\pi\overline{n}^3$ denotes the mean classical orbit time of the Rydberg electron) suddenly the excited Rydberg electron starts to diffuse to larger distances from the nucleus. Simultaneously the Rydberg electron becomes more and more spatially delocalized and $P_g(t)$ decreases significantly. This delocalization is connected with a diffusion of the Rydberg electron in energy space towards the ionization threshold. At even larger interaction times, i.e. $t \geq t_c \approx 7 \times 10^9 T$, this stochastic diffusion of the Rydberg electron in energy space reaches the ionization threshold and the ionization probability $P_{ion}(t)$ rises significantly from a negligible value to a value close to unity (compare with Fig.2a). At these times $P_g(t)$ starts to decrease significantly faster. This characteristic scenario of spatial delocalization, diffusion and stochastic ionization of a resonantly excited Rydberg electron is a characteristic phenomenon brought about by the phase fluctuations of the laser field. In particular, it implies that the diffusive dynamics of the Rydberg electron cannot be described adequately within the framework of a simple two-level approximation. Besides these qualitative aspects also the quantitative characteristics of this nontrivial long time dynamics, which are presented in Fig.2a in detail, can be worked out even analytically on the basis of the theoretical approach mentioned above. Thus it can be shown[11] that stochastic ionization starts at the critical time t_c

$$t_c = \frac{4\pi}{\gamma b\sqrt{27}}\left[\frac{(\overline{\epsilon}^2 + 3(b^2+\gamma^2/4)/4)^{3/2}}{\overline{\epsilon}^2 + b^2 + \gamma^2/4}\right]^{1/2}. \qquad (2)$$

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Furthermore, the initial state probability decays according to (green curve in Fig.2)

$$P_g(t) = \frac{(\gamma+2b)^2}{(2b\gamma)^2}\left[\frac{\gamma b\Gamma^3(5/3)}{27\pi(\overline{\epsilon}^2+b^2+\gamma^2/4)}\right]^{1/3} t^{-5/3} \qquad (t > t_c) \tag{3}$$

and the ionization probability rises according to the power law (green curve in Fig.2)

$$P_{ion}(t) = 1 - \frac{\Gamma(2/3)(\gamma+2b)}{6b\gamma}\left[\frac{\gamma b}{\pi(\overline{\epsilon}^2+b^2+\gamma^2/4)}\right]^{1/3} t^{-2/3} \qquad (t > t_c) \tag{4}$$

with $\Gamma(5/3)$ denoting the value of the Γ-function with argument 5/3. In this dynamical regime the stochastic diffusion of the Rydberg electron starts at time $t_1 = \sqrt{4/\sqrt{\pi}}/[2b\gamma T_{\overline{\epsilon}}]$ and leads to a power law decay of the form (red curve in Fig.2)

$$P_g(t) = \frac{2}{\sqrt{\pi}}[2b\gamma T_{\overline{\epsilon}} t]^{-1/2} \qquad (t_1 < t < t_c) \tag{5}$$

for the initial state probability.

Dynamical regimes in which either the bandwidth b or both the laser bandwidth b and the laser-induced depletion rate γ are larger than the mean level spacing of the excited Rydberg states are shown in Figs. 1b and 1c. Details of the associated probabilities $P_g(t)$ and $P_{ion}(t)$ are depicted in Figs.2b and 2c. In both cases the long time behaviour which is dominated by stochastic diffusion and ionization of the Rydberg electron is again described quantitatively by Eqs. (3),(4) and (5) with the stochastic ionization time t_c being given by Eq. (2). Only the initial stages of the optical excitation process prior to the stochastic diffusion are different in both cases. In Figs. 1c and 2c the bandwidth b is larger than the mean level spacing of the excited Rydberg states but the field induced depletion rate γ is still small, i.e. $\gamma T < 1$. Thus the large bandwidth b tends to smooth out the discrete energies of the Rydberg states so that the initial stage of this optical excitation process exhibits basic features of an ionization process. This smoothing process is also apparent from the wavefunction shown in Fig.1c. As a result the initial state probability decays exponentially, i.e. $P_g(t) = e^{-\gamma t}$. In Figs.1b and 2b the laser bandwith b and the depletion rate γ are both larger than the mean level spacing of the excited Rydberg states. Thus a radial electronic Rydberg wave packet is prepared by power broadening[1] whose characteristic dynamical behaviour dominates the early stages of the optical excitation process.

In order to investigate the competition between autoionization and stochastic ionization in Fig.2d the optical one-photon excitation of autoionizing Rydberg states close to an ionization threshold of an excited state of the ionic core (channel 1) is considered. In this model calculation the bandwidth b is assumed to be so large that also continuum states above this ionization threshold are located within the significantly excited energy interval $(\overline{\epsilon}-b, \overline{\epsilon}+b)$. For the sake of simplicity it is assumed that only Rydberg states in channel 1 are excited by one photon excitation from state $|g\rangle$. The early stages of this excitation process are governed by an exponential decay of the initial state $|g\rangle$ with rate γ, by autoionization of the excited Rydberg states of channel 1 into channel 2, and by direct laser-induced ionization into the continuum states of channel 1. This initial ionization process reaches a metastable equilibrium for interaction times of the order of $1/\gamma < t < t_c$. As soon as the interaction time t exceeds the stochastic ionization time t_c, this equilibrium becomes unstable and autoionization and stochastic ionization start to compete. Thus eventually a new equilibrium is established for the branching ratio between the two ionization channels. This final equilibrium is determined by the fact that all autoionizing Rydberg states whose autoionization lifetimes exceed the stochastic ionization time t_c, i.e. $1/\Gamma_n > t_c$ (Γ_n is the autoionization rate of Rydberg state $|n,1\rangle$), ionize into the continuum states of channel 1 whereas all other excited

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states autoionize into channel 2. In comparison with the metastable equilibrium in this final equilibrium autoionization may be suppressed. A neccessary condition for observing this suppression of autoionization is that there are autoionizing Rydberg states which are located within the significantly excited energy range $(\overline{\epsilon}-b, \overline{\epsilon}+b)$ and whose autoionization life times are sufficiently long, i.e. $1/\Gamma_n > t_c$. A detailed investigation of these long time phenomena which are associated with autoionization will be presented elsewhere[13].

These examples demonstrate that diffusion and stochastic ionization of a Rydberg electron are generic phenomena of optical excitation processes which originate from the destruction of quantum coherence by fluctuating laser fields. The characteristic quantitative details of these novel fluctuation-induced long time effects are described by Eqs. (3),(4) and (5). These power-laws are consequences of the basic property that Rydberg electrons can be affected by laser fluctuations only whenever they are close to the ionic core. Though the presented examples rely on idealizations, such as one and two channel approximations or the PDM for the laser fluctuations, it is expected that the quantitative details of these fluctuation-induced long time phenomena apply also to more general cases. In particular, the critical exponents appearing in Eqs. (3),(4) and (5) might be valid under quite general conditions. However, further work is needed to clarify this point.

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