Quantum Energy Flow in Atomic Ions Moving in Magnetic Fields

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(Received 12 March 1999)

Using a combination of semiclassical and recently developed wave packet propagation techniques we find the quantum self-ionization process of highly excited ions moving in magnetic fields which has its origin in the energy transfer from the center of mass to the electronic motion. It obeys a time scale orders of magnitude larger than the corresponding classical process. Importantly a quantum coherence phenomenon leading to the intermittent behavior of the ionization signal is found and analyzed. Universal properties of the ionization process are established.

PACS numbers: 32.60.+i

Rydberg atoms in strong external fields represent an exciting and very active research area both experimentally as well as theoretically. During the 1980s the main focus was on the hydrogen atom [1] assuming an infinite nuclear mass which reduces the dimensionality of the system. However, in general, the atom possesses a nonvanishing center-of-mass (CM) motion in the magnetic field, giving rise to a variety of two-body phenomena [2]. Turning to charged two-body systems like, for example, the He$^+$ ion the residual coupling of the CM and electronic motion is represented by an oscillating electric field term yielding five relevant degrees of freedom. One of the most striking effects caused by the two-body character of the ion is the recently found classical self-ionization process [3] which occurs due to energy transfer from the CM to the electronic motion. Since it is well known that quantization can severely change the effects observed in classical dynamics (see Ref. [4], and references therein) we develop in the present work a quantum approach to the moving ion in a magnetic field.

We are interested in the regime of high level density, i.e., high-lying excitations, for *both* the collective as well as the electronic motion which depend on a number of parameters (field strength, total energy consisting of the initial CM, and internal energies). The *ab initio* description of the quantum dynamics in the above regime goes even beyond modern computational possibilities and we thus seek a semiclassical approach that is capable of describing the essential physics of the problem. The total pseudomomentum [5] is a conserved quantity associated with the CM motion. In spite of the fact that its components perpendicular to the magnetic field are not independent, i.e., do not commute, it can be used to find a suitable transformation of the Hamiltonian to a particularly simple and physically appealing form [6,7]. For the He$^+$ ion it reads $\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3$, where

$$\mathcal{H}_1 = \frac{1}{2M}\left(\mathbf{P} - \frac{Q}{2}\,\mathbf{B} \times \mathbf{R}\right)^2, \tag{1}$$

$$\mathcal{H}_2 = \alpha\,\frac{e}{M}\left[\,\mathbf{B} \times \left(\mathbf{P} - \frac{Q}{2}\,\mathbf{B} \times \mathbf{R}\right)\right]\mathbf{r}, \tag{2}$$

$$\begin{aligned}\mathcal{H}_3 = {} & \frac{1}{2m}\left(\mathbf{p} - \frac{e}{2}\,\mathbf{B} \times \mathbf{r} + \frac{Q}{2}\,\frac{m^2}{M^2}\,\mathbf{B} \times \mathbf{r}\right)^2 \\ & + \frac{1}{2M_0}\left[\mathbf{p} + \left(\frac{e}{2} - \frac{Q}{2M}\,\frac{m}{M}\,(M + M_0)\right)\mathbf{B} \times \mathbf{r}\right]^2 \\ & - \frac{2e^2}{r}\,. \end{aligned} \tag{3}$$

Here, m, M_0, and M are the electron, and the nuclear and total mass, respectively. $\alpha = (M_0 + 2m)/M$ and Q is the net charge of the ion. $\mathbf{B}$ is the magnetic field vector which is assumed to point along the z axis. $(\mathbf{R}, \mathbf{P})$ and $(\mathbf{r}, \mathbf{p})$ are the canonical pairs for the CM and internal motion, respectively. The CM motion parallel to the magnetic field separates exactly and undergoes a free translational motion.

$\mathcal{H}_1$ and $\mathcal{H}_3$ depend exclusively on the CM and electronic degrees of freedom, respectively. $\mathcal{H}_1$ describes the free motion of a CM pseudoparticle with charge Q and mass M. $\mathcal{H}_3$ stands for the electronic motion in the presence of paramagnetic, diamagnetic, as well as Coulomb interactions which, in analogy to the hydrogen atom [1], exhibits a variety of classical and quantum properties with changing parameters, i.e., energy and/or field strength. $\mathcal{H}_2$ contains the coupling between the CM and electronic motion of the ion and represents a Stark term with a rapidly oscillating electric field $1/M[\mathbf{B} \times (\mathbf{P} - Q/2\mathbf{B} \times \mathbf{R})]$ determined by the dynamics of the ion. This coupling term is responsible for the effects and phenomena discussed in the present investigation.

The essential elements of our semiclassical approach are the following. Since we consider the case of a rapidly moving ion in a magnetic field a classical treatment of the CM motion coupled to the quantized electronic degrees of freedom seems appropriate: the CM is propagated with an effective Hamiltonian containing the corresponding expectation values with respect to the electronic quantum states. The latter obey a time-dependent Schrödinger equation which involves the classical CM trajectory. Both the electronic and CM motion *have to be integrated simultaneously*. The key idea of this semiclassical approach goes back to Refs. [8,9] where it has been applied to the dynamics of molecular processes. Our resulting time evolution equations read therefore as follows:

$$\begin{aligned}\frac{d}{dt}\mathbf{P}(t) &= -\frac{\partial}{\partial \mathbf{R}}\mathcal{H}_{\mathrm{cl}}(\mathbf{R}(t),\mathbf{P}(t))\\ \frac{d}{dt}\mathbf{R}(t) &= \frac{\partial}{\partial \mathbf{P}}\mathcal{H}_{\mathrm{cl}}(\mathbf{R}(t),\mathbf{P}(t)) \qquad (4)\\ i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t) &= \mathcal{H}_{q}(\mathbf{R}(t),\mathbf{P}(t),\mathbf{r})\psi(\mathbf{r},t)\end{aligned}$$

with the effective Hamiltonian

$$\begin{aligned}\mathcal{H}_{\mathrm{cl}}(\mathbf{R},\mathbf{P}) &= \mathcal{H}_1 + \langle\psi(\mathbf{r},t)|\mathcal{H}_2 + \mathcal{H}_3|\psi(\mathbf{r},t)\rangle\\ \mathcal{H}_q(\mathbf{R}(t),\mathbf{P}(t),\mathbf{r},\mathbf{p}) &= \mathcal{H}_3(\mathbf{r},\mathbf{p}) + \mathcal{H}_2(\mathbf{R}(t),\mathbf{P}(t),\mathbf{r})\,.\end{aligned} \qquad (5)$$

This scheme represents a balanced treatment of the coupled classical and quantum degrees of freedom of the ion and takes account of the energy flow among them. It possesses the important property of conserving the total energy which is particularly important for the correct description of the energy transfer processes occurring in our system. Since the typical energies associated with the fast heavy CM degrees of freedom are many orders of magnitude larger than the corresponding elementary quantum ($\hbar QB/M$) we expect the above scheme to yield reliable results.

Our approach to the solution of the time-dependent Schrödinger equation, which yields the dynamics of an initially defined wave packet $\psi(\mathbf{r},t)$, is based on a recently developed nonperturbative hybrid method [10–12]. It uses a global basis on a subspace grid for the angular variables (θ,ϕ) and a variable-step finite-difference approximation for the radial variable. The angular grid is obtained from the nodes of the corresponding Gaussian quadrature with respect to θ and ϕ, which is in the spirit of the discrete variable techniques yielding a diagonal representation for any local interaction [10]. As a consequence one remains with the Schrödinger-type time-dependent radial equations coupled only through nondiagonal matrix elements of the kinetic energy operator. This vector equation is propagated using a splitting-up method [13], which permits a simple diagonalization procedure for the remaining nondiagonal part [11,12]. Our scheme is unconditionally stable, saves unitarity, and has the same order of accuracy as the conventional Crank-Nickolson algorithm, i.e., $\sim O(\Delta t^2)$, where Δt is the time step size. In order to avoid reflections of the wave packet from the right edge of the radial grid we introduce absorbing boundary conditions. The extension of the radial grid is chosen (see below) such that it exceeds the center of the radial distribution of the initial wave packet by more than 1 order of magnitude. The typical frequencies associated with the motion of the Rydberg electron and the CM motion are different by several orders of magnitude (see Ref. [3]). To investigate the quantum energy transfer mechanisms requires therefore the integration of the above equations of motion for a typical time which is a multiple of the time scale of the heavy particle (CM). This corresponds to many thousand cycles of the Rydberg electron. Such a detailed investigation would have been impossible without the use of the above-described combination of highly efficient techniques.

We assume that the He$^+$ ion is accelerated up to some value E_{CM} of the kinetic energy of the CM motion and its electron is being excited to some Rydberg state nlm in field-free space. Thereafter it enters the magnetic field. In the following we choose $E_{\mathrm{CM}} = 100$ a.u., $n = 25, l = m = 0$, and a strong laboratory field $B = 10^{-4}$ a.u. (23.5 T). The initial CM velocity is $v_{\mathrm{CM}} = 0.1656$ a.u. and oriented along the x axis. We remark that taking the above values for nlm, B for the He$^+$ ion with the assumption of an infinitely heavy nucleus the corresponding classical phase space is dominated by chaotic trajectories. Figures 1(a) and 1(b) illustrate results

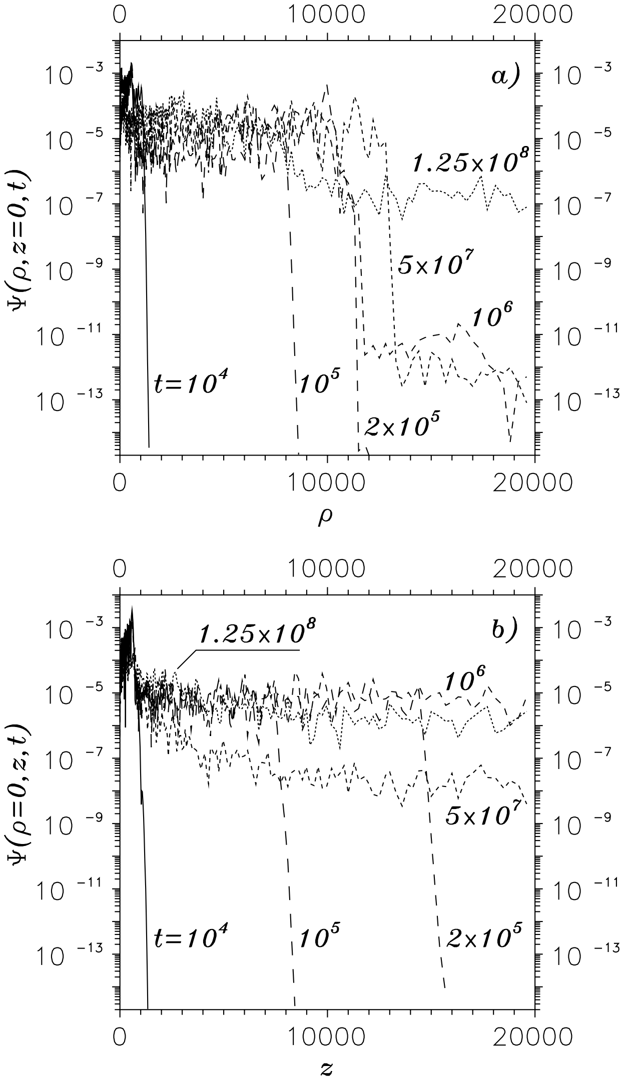

FIG. 1. (a) The intersection $\Psi(\rho, z = 0, t) = \int|\psi|^2\,d\phi$ along the ρ axis. (b) The intersection $\Psi(\rho = 0, z, t)$ along the z axis. (Atomic units are used.)

for the propagation of the wave packet with increasing time. More precisely we show the intersection of the integrated quantity $\Psi(\rho, z, t) = \int |\psi|^2 \, d\phi$ along the cylindrical ρ axis for $z = 0$ (Fig. 1a) and its intersection along the z axis for $\rho = 0$ (Fig. 1b). Figure 1a demonstrates that the motion of the wave packet is confined by the diamagnetic interaction with respect to the ρ direction, i.e., the direction perpendicular to the magnetic field. For any propagation time its value drops by several orders of magnitude at some outer value ρ_c for the ρ coordinate. As we shall see below (see also Fig. 3) the variation of ρ_c is accompanied by a corresponding change in the internal/CM energies thereby demonstrating the flow of energy between the CM and electronic degrees of freedom. Figure 1b demonstrates that there is for certain time intervals (see below) almost no decay of the wave packet for large distances of the z coordinate. Therefore we encounter a significant flux of probability parallel to the external field. Reaching the boundary of our radial grid $r_m = 20\,000$ a.u. it is absorbed and considered to represent the ionized state. Having established the existence of an ionizing probability flux parallel to the magnetic field we immediately realize from Fig. 1b that this flux is by no means constant in time but varies strongly. To see this more explicitly and also to gain an idea of the overall decay of the wave packet we show in Fig. 2 the decay of the norm of the wave packet for a time which roughly corresponds to one cyclotron period $2\pi M/(QB)$ of the free CM motion in the field. Figure 2 shows apart from an overall monotonous decay of the norm, which is due to the quantum self-ionization process, an amazing new feature: the norm exhibits an alternating sequence of plateaus and phases of strong decay. The widths of the plateaus slightly increase with increasing time. This intermittent behavior of the ionization signal from the moving ions is a pure quantum phenomenon, i.e., does not occur in the corresponding classical ionization rates [3]. Furthermore the calculation of the classical ionization rates for the same parameters (field strength, energies) yields a typical ionization time which is by 2 orders of magnitude smaller than the one obtained by the quantum calculation. The ionization process is therefore significantly slowed down through the quantization of the system which is in the spirit of the quantum localization processes shown to exist in a variety of different physical systems (see Ref. [4], and references therein). The observed slowing down of the ionization process represents one important difference of the classical and quantum behavior of the moving ion which occurs in spite of the fact that we are dealing with a highly excited system.

The obvious question of the origin of the intermittent occurrence of the plateaus and ionization bursts arises now. At this point it is helpful to consider the behavior of the CM energy as a function of time which is illustrated in Fig. 3. Starting with $E_{\mathrm{CM}} = 100$ a.u. at $t = 0$ we observe a fast drop of it for short times yielding a minimum of E_{CM} at approximately $t_1 = 4 \times 10^7$ a.u. Thereafter it raises and reaches a maximum at approximately $t_2 = 1.2 \times 10^8$ a.u. after which it drops again; i.e., it shows an overall oscillating behavior. The "valleys" of E_{CM} coincide with the plateaus of the norm decay whereas the regions with higher CM velocities correspond to phases of a strong norm decay of the wave packet. The increase of the widths of the plateaus in the norm decay (see Fig. 2) matches the corresponding decrease of the frequency of the oscillations of the CM energy. Since the total energy is conserved this clearly shows that the ionization bursts correspond to phases of relatively low internal energy (although certainly above the ionization threshold) whereas the phases of higher internal energy go along with the plateaus of

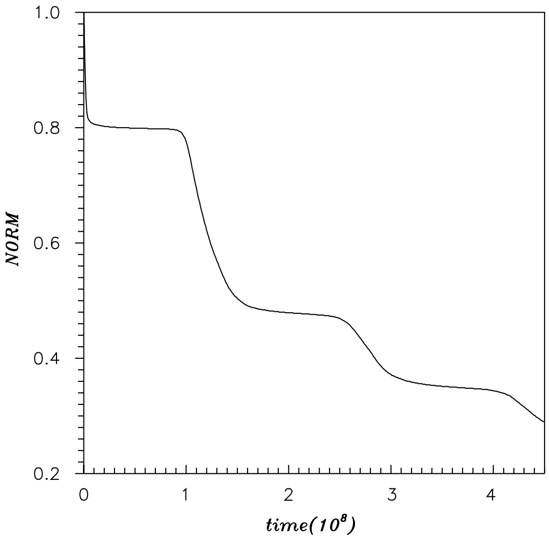

FIG. 2. The norm of the electronic wave packet as a function of time (in units of 10^8 atomic units).

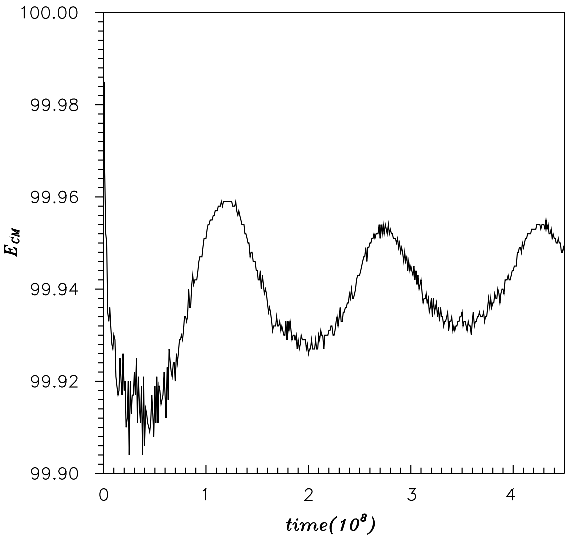

FIG. 3. The CM energy as a function of time (in units of 10^8 atomic units).

the norm behavior, i.e., the localization of the electronic motion. This provides the key for the understanding of the rich structure of the norm decay. The phase of high energy for the electronic motion means that the magnetic interaction strongly dominates over the Coulomb interaction. This makes the electronic motion approximately separable with respect to the motion perpendicular and parallel to the magnetic field. As a consequence the energy transfer process from the degrees of freedom perpendicular to those parallel to the external field are very weak; i.e., the ionization process is strongly suppressed. This corresponds to an almost integrable situation for the ions dynamics. On the contrary for relatively low internal energies the Coulomb interaction is much more relevant and mediates together with the coupling Hamiltonian $\mathcal{H}_2$ the energy transfer from the CM to the electron motion parallel to the magnetic field. As a result we encounter a flow of probability in the $+/-$ z directions which corresponds to the ionization burst. During this period of motion a comparatively strong dephasing of the wave packet takes place.

The intermittent behavior of the ionization rate can therefore be seen as a quantum manifestation for the switching between different regimes of the internal energy corresponding to weaker or stronger Coulomb interaction. Pumping energy from the CM to the electronic motion weakens the Coulomb interaction and leads to the suppression of the ionization process whereas pushing the energy back to the CM motion decreases the internal energy and enhances the Coulomb interaction. To elucidate the time scale on which this process takes place we have computed the autocorrelation function $C(t) = \langle \psi(t) \mid \psi(0) \rangle$ where $\langle\,\rangle$ means integration over the electronic coordinates. As a result we observe a modulation and recurrence of the autocorrelation at a time scale $t \approx 1.6 \times 10^8$ a.u. which corresponds approximately to the recurrence of the plateaus for the norm decay. The corresponding power spectrum shows a broad peak at a frequency $\omega \approx 3.5 \times 10^{-8}$ a.u. An important feature of the quantum self-ionization process is the approximate stability of the time intervals corresponding to the plateaus of the norm (no ionization signal) with respect to variations of the initial CM velocity of the ion. Our investigation shows that decreasing the CM energy from 100 to 12 a.u. leads to a decrease with respect to the distances between the plateaus, i.e., the difference of the norm values belonging to different plateaus, roughly by a factor of 2. This corresponds to a significant slowing down of the ionization process. However, the widths of the plateaus remain rather stable and *represent therefore a universal quantity* which is approximately independent of the CM velocity. Varying the field strength causes a change of both the distances between the plateaus and their widths.

The quantum self-ionization process should have implications on the physics of atoms and plasmas occurring in a number of different circumstances. Apart from this it obviously suggests itself for a laboratory experiment (the lifetime of the Rydberg states exceeds the time scale of ionization by orders of magnitude) which should be very attractive due to the expected intermittent ionization signal which is a process revealing the intrinsic structure and dynamics of the system during its different phases of motion.

This work was supported by the National Science Foundation through a grant (P. S.) for the Institute for Theoretical Atomic and Molecular Physics at Harvard University and Smithsonian Astrophysical Observatory. P. S. thanks H. D. Meyer and D. Leitner for fruitful discussions. V. S. M. gratefully acknowledges the use of the computer resources of the IMEP of the Austrian Academy of Sciences; he also thanks the PNTPM group of the Université Libre de Bruxelles for warm hospitality and support.

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