Detecting classical bifurcations in atoms from experimental data

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This Rapid Communication proposes an experiment with Rydberg atoms resolved simultaneously in energy and time. The occurrence of classical bifurcations can be detected directly from experimental data and, therefore, information about the nonintegrability of classical equations of motion can be obtained. Results for the hydrogen atom subjected to a static uniform magnetic field are presented and discussed in detail.

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In the study of the interrelation between classical nonintegrable systems and quantum systems, atoms subjected to external magnetic fields have received special attention in the last years. The role of classical periodic orbits, in the external magnetic fields have received special attention in integrable systems and quantum systems, atoms subjected to

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possibility that satisfies the first step, and that will be used in this Rapid Communication, is the creation of electronic radial Rydberg wave packets with short laser pulses (for a comprehensive review, see [7]).

If a short laser pulse (~ps or fs) with polarization $e_1$ and frequency $\omega_1$ excites an electron from a low-lying initial $|g\rangle$ state to a coherent superposition of many Rydberg states, then a nonstationary electronic radial Rydberg wave packet (measurable quantity) is "created." This wave packet, which is localized in the radial coordinate, is centered around the mean excited energy $\bar{\epsilon}(\text{a.u.})=-1/(2 \bar{r}^2)$, where $\bar{r}$ is the effective principal quantum number. The wave packet moves periodically between two turning points, one near the core and the other far from it. The period of oscillation of the wave packet between the two turning points agrees with the semiclassical return time $T_R(\text{a.u.})=2\pi \bar{r}^3$ of a classical orbit with energy $\bar{\epsilon}$. If the pulse time of the pump laser is shorter than the classical orbit time, then the wave packet is localized in time. From the experimental point of view, time resolution is obtained when the evolution of the wave packet can be "observed." This can be done with a short probe laser, with polarization $e_2$ and frequency $\omega_2$, which deexcites the electron by stimulated emission of a photon to a low-lying final $|f\rangle$ state. The excited electron can absorb the photon from the probe laser only when the wave packet is localized near the core. If the frequency of both pump and probe lasers is kept fixed and the time delay $\Delta t$ between them is varied, it is possible to study the time evolution of the wave packet. Technically, this time evolution consists in computation of the two-photon transition amplitude [7],

$$P_{fg} = \left| \frac{1}{2\pi} \int_{-\infty}^{\infty} d\epsilon e^{-i\epsilon(t_2-t_1)} T_{fg}(\epsilon) \bar{\epsilon}_1(\epsilon-\bar{\epsilon}) \bar{\epsilon}^*_2(\epsilon-\bar{\epsilon}) \right|^2,$$

between an initial state $|g\rangle$ and a final state $|f\rangle$ as a function of the time delay $\Delta t=t_2-t_1$ between pump and probe lasers. The two-photon transition amplitude $T_{fg}(\epsilon)$ that appears in Eq. (1) is given by

$$T_{fg}(\epsilon) = \langle f| d \cdot e_2^* (\epsilon+i0-H)^{-1} d \cdot e_1 |g\rangle,$$

where $H$ is the Hamiltonian operator of the system in the absence of the lasers, $d$ is the dipole operator, and

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In order to analyze the behavior of the wave packet when the pump-probe experiment is repeated, it is convenient to take a time delay between the two lasers that corresponds to a maximum in the experimental signal. In other words, a maximum of the return time of the wave packet to the core is kept fixed, while the frequency (i.e., the mean excited electron energy) and the time delay between pump and probe lasers are varied. To maintain fixed, it is necessary that the relation between the mean excited electron energy and the time delay always satisfy the relation $\Delta t = nT_{\bar{\nu}}$. Therefore, if the energy and $\Delta t$ were varied simultaneously, in conformity with the above relation, the dependence of the $n$th return of the wave packet to the core could be studied as a function of the energy. In other words, the maximum (for a given $n$ of Fig. 1, for example) of the transition probability $P_{fs}$ could be studied as a function of the electron energy.

Figure 2 shows the energy-tuned pump-probe experiment for the case of the hydrogen atom without external fields. The transition probabilities of the three first maxima of Fig. 1 (i.e., $n=1,2,3$) are shown as a simultaneous function of the quantum number and time, i.e., $\bar{\nu}, \Delta t$. When the energy goes to zero ($\bar{\nu} \rightarrow \infty$), the curves decay with a rate $[n\bar{\nu}^2]^{-1/2}$ [7]. For the quantum number $\bar{\nu} = 80 (\Delta t = 77 \text{ ps})$, the wave packets of Fig. 2 are the same as in Fig. 1. Note that, although the three curves in Fig. 2 have been superposed, the experiment is realized separately for each return $n$. In the limit for energies $\bar{\nu} \rightarrow 0 (\bar{\nu} \rightarrow \infty)$ and longer return times of the wave packet to the core (i.e., near the classical escape energy $\bar{\nu} = 0$, right-hand side of Fig. 2), more Rydberg states are excited and the wave packet is better localized. This limit is called the classical limit, while the other extreme (lower energies and shorter return times) is called the quantum limit, when the localization of the wave packet is lost.

The energy-tuned pump-probe experiment will now be discussed for a nonintegrable system: the case of a hydrogen atom subjected to a static uniform magnetic field in the $e_\parallel$ direction. In this case, the transition amplitude (2) can be described in the isolated-orbits approximation by [7]

\[
\bar{E}_k(t) = \int_{-\infty}^{\infty} dt e^{i\omega t} \bar{E}_k(t + t_i) \quad (k=1,2)
\]

where $\bar{E}_k(t)$ are the Fourier transforms of the laser pulses' envelopes that are described by classical electric fields. The main problem in this theoretical description is the determination of the energy dependence of the two-photon transition amplitude $T_{fs}(\bar{\nu})$.

To illustrate what happens in the "classical" pump-probe experiment, Fig. 1 shows an example from the periodic motion of a wave packet for the case of a hydrogen atom without external field. The initial and final states are $|s\rangle$ states, while the intermediate Rydberg states are $|p\rangle$ states. Maxima in the transition probability of Fig. 1 are always observed when the time delay is given by $\Delta t = nT_{\bar{\nu}}$ ($n=1,2,3,\ldots$), i.e., corresponds to the $n$th return of the wave packet to the core. With the help of quantum-defect theories and semiclassical methods, the transition amplitude (2) can be written as a sum over classical periodic orbits starting from the core and returning to it with a pure radial impulse [7].

In this semiclassical description of wave packets it has been shown [7] that the transition probability (1) always has maxima when the time delay between the two lasers is a multiple of the classical return time of the electron. In the case of the hydrogen atom without external fields, it is possible to calculate semiclassically an analytical expression of the transition probability (1) [see Eq. (41) of [7]].

In the second part of the experiment resolved simultaneously in energy and time, the mean excited energy $\bar{\epsilon}$ of the center of the wave packet must be changed. This can be accomplished by changing the laser frequency and, in consequence, the wave packet is centered around another Rydberg state. The period of oscillation of the wave packet is changed in conformity with the relation $T_{\bar{\nu}} = 2\pi \bar{\nu}$. In order to analyze the behavior of the wave packet when the energy is changed, it is convenient to take a time delay between the two lasers that corresponds to a maximum in the transition probability (i.e., $\Delta t = nT_{\bar{\nu}}$; see maxima of Fig. 1). In this case the observed experimental signal is stronger. Since for each new laser frequency the return time of the wave packet to the core is also changed, it is necessary that $\Delta t$ be varied simultaneously in order to obtain again a maximum in the experimental signal. In other words, a maximum of the $n$th return of the wave packet to the core is kept fixed, while the frequency (i.e., the mean excited electron energy) and the time delay $\Delta t$ between pump and probe lasers are varied. To maintain fixed, it is necessary that the relation between the mean excited electron energy and the time delay always satisfy the relation $\Delta t = nT_{\bar{\nu}}$. Therefore, if the energy and $\Delta t$ were varied simultaneously, in conformity with the above relation, the dependence of the $n$th return of the wave packet to the core could be studied as a function of the energy. In other words, the maximum (for a given $n$ of Fig. 1, for example) of the transition probability $P_{fs}$ could be studied as a function of the electron energy.

FIG. 1. Transition probability (in arbitrary units) vs time delay (in units of the classical orbit time $T_{\bar{\nu}} = 77 \text{ ps}$, corresponding to $\bar{\nu} = 80$). The pulse time of the laser is $\tau = 8.5 \text{ ps}$.

\[ P_{ss}(\Delta t) = \int_{-\infty}^{\infty} dt e^{i\omega t} P_{ss}(t + t_i) \]

\[ \bar{E}_k(t) = \int_{-\infty}^{\infty} dt e^{i\omega t} \bar{E}_k(t + t_i) \quad (k=1,2) \]

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where $\varepsilon_m = \varepsilon + m \gamma$. The behavior of the two-photon transition amplitude $T_{fs}(\varepsilon)$ is characterized by the property of classical quantities like the action $S_j$, the Maslov index $\mu_j$, the number of crossings ($\nu_j$) of the axis $\varepsilon$, the initial ($\theta_j$) and final $\theta_j$ angles of the orbits, the classical cross section

$$
\sigma_j^{(n_j)}(\varepsilon) = \frac{\sin \theta_j \sin \theta_{oj}}{\partial \theta_j \sin \mu_j},
$$

as well as the photoionization $\mathcal{D}_{lm}^{(+)}$ and photorecombination $\mathcal{D}_{lm}^{(-)}$ dipole matrix elements. The index $j$ represents the isolated orbits, while $\mu_j$ is the stability index of the orbits and $p_{\theta}$ is the canonical momentum associated with $\theta$. The number $n_j$ represents the number of returns of the orbit $j$ to the core. The term $T_{fs}^{(i)}$ in Eq. (3), represents a direct part of the transition amplitude, while the terms with a summation over $j$ represent the contribution of all excited classical orbits.

Figure 3 shows the transition probability $P_{fs}$ between an initial $|s\rangle$ and a final $|s\rangle$ state, for the returns $n = 1, 2, 3$ of a radial Rydberg wave packet to the core, as a simultaneous function of the scaled time $\gamma \Delta t$ and the scaled energy $\tilde{\varepsilon} = \varepsilon / \gamma^{\alpha_3}$, where $\gamma$ represents the magnetic-field intensity in atomic units given by $B_0 = 4.71 \times 10^5$ T. The intermediate Rydberg states are $|p\rangle$ states. The time delays between pump and probe lasers were chosen to correspond exactly to the return of the center of the wave packet to the core. For $n = 1, 2, 3$, the time delays used were, respectively, $\Delta t = T_{fs}, 2T_{fs}$, and $3T_{fs}$. The laser predominantly excites the orbit perpendicular to the magnetic field (represented here by $I_0$ and with the index $j = 0$), the pulse time is of the order of 0.9 ns, and the excited energy is varied between $-1.5 \text{ (a.u.)} \leq \varepsilon \leq -0.1 \text{ (a.u.)}$, which corresponds to a change in the time delay from 23 ns to $\Delta t \leq 63$ ns. To understand the results and the accentuated peaks that appear in Fig. 3, classical properties of the electronic motion are briefly summarized in what follows.

It is known [2,14], that the classical motion of an electron subjected simultaneously to a Coulomb potential and a magnetic field, presents different behavior for different scaled energies. In the limit $\tilde{\varepsilon} \to -\infty$, called the integrable limit, the motion is regular. In the opposite limit, when the energy approaches the escape energy $\tilde{\varepsilon} = 0$, the motion is totally irregular (chaotic) and all orbits are unstable. Between these two limits, for energies $-1.0 \leq \tilde{\varepsilon} \leq -0.2$, there is a mixing regime of regular and irregular motion and bifurcation phenomena can occur, inducing a proliferation of orbits when the energy approaches $\tilde{\varepsilon} = -0.2$. In particular, for the perpendicular orbit $I_0$, a period-1 bifurcation takes place for the energy $\tilde{\varepsilon}_1 \approx -0.2028$ and becomes unstable. For the second return ($n_0 = 2$) of $I_0$ to the core, there is a period-4 bifurcation for the energy $\tilde{\varepsilon}_2 \approx -0.50191$. For $n_0 = 3$, a bifurcation of period-6 occurs for $\tilde{\varepsilon}_3 \approx -0.7664$ and a period-3 bifurcation for $\tilde{\varepsilon}_3 \approx -0.3322$.

In the example of Fig. 3, the energy-tuned pump-probe experiment is simulated for energies that include a classical motion that goes from regular up to chaotic. The energy dependence of the second return ($n_0 = n = 2$; see dotted curve in Fig. 3) of the wave packet, which moves around the orbit $I_0$, has an accentuated maximum near the energy $\tilde{\varepsilon}_2 \approx -0.50191$, where a classical bifurcation occurs. This maximum is due to the singularity of the cross section (4) and reflects the physical situation that near classical bifurcation energies other new orbits $I_{z+1}, I_{z+2}$ (as, for example, those in Fig. 2 from [8]) appear near $I_0$. This also means that in the excited energy range, a “focusing” effect occurs, when a large number of closed orbits return to the core. The experimental signal observed in the transition probability is therefore considerably stronger.

Despite the fact that, near classical bifurcations, the cross section (4) diverges and the isolated orbits approximation (3) is no longer correct, the transition amplitude $T_{fs}(\varepsilon)$ has been derived in a uniform approximation that is valid near the above-mentioned bifurcation (for details, see [8]). The minimum, which appears to the right-hand side of the $n = 2$ peak, is due to a destructive interference between those fractions of the wave packet that returned along the stable orbit $I_0$ and those fractions that returned along the unstable orbits $I_{z+2}$. Maxima of the same type which appeared in the curve for $n = 2$, can be also observed near the bifurcations $\tilde{\varepsilon}_{31}$ and $\tilde{\varepsilon}_{32}$, when the third return of the wave packet to the core is considered (see dashed curve in Fig. 3, $n_0 = n = 3$). For the solid line of Fig. 3 ($n_0 = n = 1$), no maximum can be observed near the classical bifurcation $\tilde{\varepsilon}_1$, since in this case Eq.
(4) does not diverge. This bifurcation phenomenon cannot be observed in the energy-tuned pump-probe experiment, since it does not occur near the core [14].

From the existence of maxima in Fig. 3, one may conclude just from experimental data, that classical bifurcations occur and, therefore, that the classical system is nonintegrable. The experimental values of the energies where maxima occur, provide good approximations of the localization of classical bifurcations for the second and third returns of the orbit \(I_0\) to the core.

Due to scaling properties of the Hamiltonian of the hydrogen atom in a static magnetic field (see [2]), the proposed experiment could be performed by changing the magnetic field intensity instead of the frequency of the laser.

To conclude, in this paper an energy-time resolved experiment has been proposed and discussed semiclassically. This experiment is ideally suited to study the effects of classical bifurcations for the second and third returns of the orbit perpendicular to the magnetic field was analyzed. At the classical bifurcation energies, the proposed experiment shows peaks due to the singularities in the classical cross section. These peaks do not appear in the integrable case.

It would be interesting to perform an energy-tuned pump-probe experiment for hydrogenic and nonhydrogenic atoms in external fields, where models of the core potential were used, in an attempt to describe scattering effects in the classical dynamics [3,5]. Since the proposed experiment would give approximately the energy where classical bifurcation is expected to take place, the method proposed seems ideally suited for studies of novel bifurcation phenomena due to core scattering effects. It might also be of value in assessing the goodness of the core potential models.

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