Photoabsorption dynamics of doubly excited two-electron systems: Closed classical orbits

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We describe how closed-orbit theory can be applied to the photoabsorption dynamics of asymmetric doubly excited atoms, and demonstrate the method using He and Yb. Closed classical orbits are found to exist and are explained in terms of electron correlation and core scattering. In He, where long range electron correlation alone influences the orbits, the results give insight into the role of electron correlation on closed classical orbits.

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Semiclassical closed-orbit theory (COT) has achieved great success for the spectroscopy of atomic singly excited Rydberg states in external fields [1–5]. For other atomic chaotic systems, such as the three-body Coulomb system, the borderline between classical and quantum mechanics is largely vague, with a complete semiclassical approach still not available due to electron correlation. Electron correlation plays a significant role, not only in atomic physics, but also in solid-state physics and may hold the key to the development of quantum dots as future electronic switches. Consequently, correlated two-electron dynamics is a vigorous research area with a proper description still an outstanding challenge. We present here a study of correlated two-electron dynamics using semiclassical closed-orbit theory. We apply this closed-orbit theory and recurrence spectroscopy to systems in which electron correlation is present.

For helium double-Rydberg states (DRS), early semiclassical approaches used simple periodic orbits to predict resonance structure in the DRS excitation spectrum [6], which are either unstable (Wannier orbits) or not reachable (Langmuir orbits) with current experimental techniques, and, therefore, do not help to understand the observed spectra. A more complicated type of stable orbit, the ''frozen-planet'' orbit [7], has been investigated extensively. However, there is little convincing experimental evidence connected to these orbits, although their discovery was promoted by experiment [7,8].

In this article, we present a semiclassical approach developed from COT for the electron dynamics in laser excitation of asymmetric DRS and use it to calculate the recurrence spectrum for He and Yb. Long-range electron-electron correlation is taken into account through the classical equations of motion. Quantum defects are incorporated via classical scattering of orbits. We also present the recurrence spectrum of DRS from the experiment, obtained by analyzing a previously measured Yb absorption spectrum [9]. Recurrences are classical electron orbits that leave from an atom and later return. Overall agreement between our calculated results and the experimental recurrence spectrum demonstrates that our model captures most of the main dynamics features of the experiment and reveals that COT can describe the effects of long-range dielectronic correlation and quantum defects in DRS excitation.

An asymmetric DRS is prepared experimentally using the isolated-core excitation technique, where the outer electron is a spectator in a Rydberg orbit while the inner electron is excited by laser photons into a lower Rydberg orbit. The interaction between the outer electron and the laser field is neglected. The semiclassical dynamics is investigated and applied to the inner electron using COT by replacing the usual static-applied external field with a time-dependent field provided by the outer electron, as summarized below. COT with time-dependent external fields has been applied to singly excited Rydberg states by Spellmeyer *et al.* and recently formally developed by Haggerty and Delos [4].

COT describes Rydberg excitation in the following way. When the inner electron is excited by the laser field, it goes into an outgoing wave propagating away from the atomic core. Sufficiently far from the core, the wave propagates according to classical mechanics, and is correlated with classical trajectories. Some of the trajectories will eventually be turned back to the vicinity of the core, and the associated incoming wave interferes with the outgoing wave to produce oscillations in the absorption spectrum. These trajectories are called closed orbits. A recurrence spectrum can be obtained by Fourier transform of the absorption spectrum. In the recurrence spectrum, each peak is located at the classical action of a closed orbit. Peak heights are related to the classical amplitudes of the closed orbits associated with the divergence of the returning trajectories in the vicinity of the core.

In our approach, the outer electron is treated as a classical particle in the excitation process of the inner electron, and together with the core, provides a time-dependent potential for the inner electron. The outer electron itself is moving in a Rydberg orbit when the inner electron is launched, and consequently will produce different results at different initial positions, which are weight averaged in our calculation. Our approach focuses on the classical dynamics of the trajectories. We do not include quantum-interference effects, exchange effects, or quantum dynamics in the vicinity of the core. We incorporate the core using classical scattering.

To produce a recurrence spectrum from experiment requires a background-free absorption spectrum [9,10], which for DRS is available for Yb. We will first describe the theoretical approach for two-electron He, where the inner electron and the core form a hydrogenic system, and then we will incorporate the influence of the core for Yb.

The classical action and the amplitude for each closed orbit can be calculated based on the Hamilton equations of

motion. The classical Hamiltonian for a two-electron atomic system with infinite mass can be written as

$$H = p_1^2/2 + p_2^2/2 - Z/r_1 - Z/r_2 + 1/r_{12}, \tag{1}$$

where p_1, p_2 and r_1, r_2 denote the inner electron and the outer electron momentum and distance from the nucleus, respectively, and the distance between the electrons is r_{12}. (Atomic units are used unless specified.) We label the excited DRS with $NLnl$ using the independent electron model, where NL and nl denote the initial principle and angular momentum quantum numbers of the inner and outer electron respectively, with the inner electron launched near the core into a Rydberg orbit by the laser field. To compare the calculated and experimental results, we use scaled coordinates defined as

$$\tilde{r}_i = Z r_i / N^{*2}, \quad i = 1,2,12;$$

$$\tilde{p}_j = N^* p_j / Z, \quad j = 1,2. \tag{2}$$

Here the asterisk denotes the effective quantum number. The scaled Hamiltonian can be written as

$$\tilde{H} = \frac{N^{*2}}{Z^2} H = \frac{\tilde{p}_1^2}{2} + \frac{\tilde{p}_2^2}{2} - \frac{1}{\tilde{r}_1} - \frac{1}{\tilde{r}_2} + \frac{1}{Z\tilde{r}_{12}}. \tag{3}$$

This scale transformation does not change the classical dynamics of the system, which can be characterized by the scaled total energy (or equivalently, by N^*/n^*)

$$\varepsilon = \frac{N^{*2}}{Z^2} E = -\frac{1}{2}\left[1 + \left(\frac{Z-1}{Z}\right)^2 \left(\frac{N^*}{n^*}\right)^2\right], \tag{4}$$

where E is the unscaled total energy. A system with different E but the same ε maintains a constant phase-space structure, keeping the classical dynamics and the set of closed orbits unchanged.

Due to the presence of the outer electron, the inner-electron energy is not constant. To calculate the classical action for the closed orbit of the inner electron in the extended phase space, we introduce the time-dependent Hamiltonian for the inner electron, expressed in scaled variables

$$\tilde{H}_t = \frac{N^{*2}}{Z^2} H_t = \frac{\tilde{p}_1^2}{2} - \frac{1}{\tilde{r}_1} + \frac{1}{Z\tilde{r}_{12}}. \tag{5}$$

The Hamiltonian has the form of a perturbed Kepler problem. The scaled generalized classical action (in units of 2π) of the closed orbit is

$$\tilde{S}_1 = \frac{1}{2\pi N^*} S_1 = \frac{1}{2\pi} \oint \{\tilde{\mathbf{p}}_1 \cdot d\tilde{\mathbf{q}}_1 - [\tilde{H}_t(t) - \varepsilon_{ti}] d\tilde{t}\}, \tag{6}$$

where ε_{ti} is the initial value of $\tilde{H}_t$ when the orbit is launched.

Using the key result of COT that closed orbits produce sinusoidal variations in the absorption rate as a function of the energy, the semiclassical prediction for the oscillator strength density observed in the absorption spectrum is

$$Df(N^*,\varepsilon) = Df_0(N^*) + \sum_k C_k(N^*) A_k(\varepsilon) \times \sin(2\pi \tilde{S}_{1k} N^* + \Delta_k), \tag{7}$$

where k labels all closed orbits and their repetitions. Here Df_0 and C_k, determined by the initial state and the laser properties (e.g., polarization, intensity, and photon energy), vary slowly with N^*, and are approximated as constant here. $A_k(\varepsilon)$ is the classical amplitude of the kth closed orbit and is a function of ε. The phase Δ_k includes the Maslov index term and a phase shift.

In COT the classical amplitude $A_k(\varepsilon)$ is associated with the divergence of the closed orbit. The standard procedure uses the Jacobian matrices to calculate $A_k(\varepsilon)$, which can require complicated mathematics in some situations, for example, chaotic trajectories [2]. We have developed an approximate method to evaluate $A_k(\varepsilon)$ without the need to calculate the matrices. Briefly, a bundle of orbits around a closed orbit [11,12] is launched from the core. An orbit in the bundle is regarded as quasiclosed according to a ''quantized'' rule that upon return to the vicinity of the core, the change in its angular momentum L and projection L_z at the inner turning point is less than ~ 0.5 a.u. (The precise value chosen is not important.) $A_k(\varepsilon)$ is directly related to the number of quasiclosed orbits in the bundle, i.e., the more quasiclosed orbits, the larger $A_k(\varepsilon)$. Detailed description will be given elsewhere.

In order to find the closed orbits of the inner electron, we launch a p electron from the core and propagate the classical three-dimensional trajectories of both electrons applying the Hamiltonian in Eq. (3). A 50-node Beowulf cluster computer in the Wesleyan Physics Department is used for the calculations. Brute force application of the Hamilton-Jacobi equations cannot be expected to yield accurate solutions due to a combination of complications including high dimensionality, singular potentials, and long-range interactions. Our calculation is not intended to possess the rigor necessary to describe complex details of DRS dynamics but rather to reproduce the important features of the experimental recurrence spectra, thereby gaining insight into the properties of the classical closed orbits that compose it, and thus revealing the physical origin of the spectra in terms of electron correlation and core scattering.

Experimentally, the recurrence spectrum [the spectrum of $|C_k(N^*)A_k(\varepsilon)|$ as a function of $\tilde{S}_1$], is obtained by performing the Fourier transform of $[Df(N^*,\varepsilon) - Df_0(N^*)]$ as a function of N^*, giving the relative amplitudes and actions of closed orbits. An ideal absorption spectrum is the ''scaled spectrum'' where the scaled orbital periods are constant over the observation interval thereby providing sharp resonances. In the present case this means that N^*/n^* or ε is kept constant. However, for the experimental spectrum n^* is constant while N^* is scanned. This means that the classical amplitude $A_k(\varepsilon)$ in Eq. (7) is changing. Nonetheless, since N^*/n^* does not change significantly, $A_k(\varepsilon)$ can be regarded as constant,

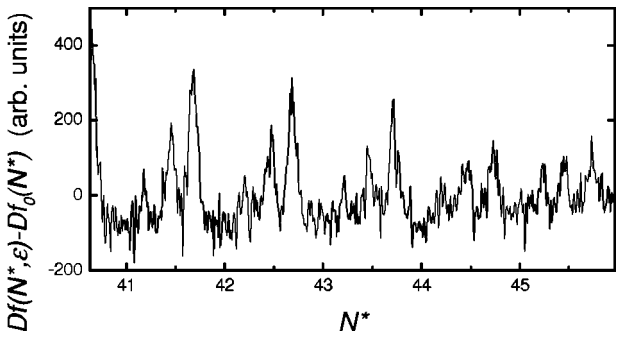

FIG. 1. Experimental absorption spectrum [9] of Yb DRS $Np80d$ ($N^*=40.63$–$45.97, n^*=77.3$) obtained by laser excitation from $7s80d$.

which makes the Fourier transform of the absorption spectrum at fixed n^* a very good approximation to the scaled recurrence spectrum.

Figure 1 shows the experimental absorption spectrum reported in Ref. [9] plotted here versus N^* rather than the laser frequency. The corresponding recurrence spectrum is shown in Fig. 2(a). Recurrence spectrum of another experimental absorption spectrum with the same $\bar{\varepsilon}$, but different $N^*(38$–$43), n^*(72.3)$ shows nearly identical structure, indicating that the system has well-defined closed orbits that determine the structure of the absorption spectrum.

In our theoretical simulation we consider the case of He, where the inner electron is hydrogenic and core effects are not present. Figure 2(b) shows our calculated He DRS recurrence spectrum. The spectrum is dominated by the primitive orbit with its repetitions present but greatly reduced in intensity. The spectrum is not that of field-free hydrogen owing to the long-range electron correlation. For asymmetric DRS, long-range electron correlation modifies slightly the classical trajectories such that the primitive orbit repetitions, which are not exactly those of standard COT due to the electron correlation, are diminished. The primitive recurrence peak is composed of about 80% eZe orbits (the inner electron is localized away from the outer electron) and about 20 % Zee orbits (localized toward the outer electron). For vanishing electron interaction, i.e., in the limit of very large n^*, the spectrum becomes that of field-free hydrogen with large repetition peaks of equal height.

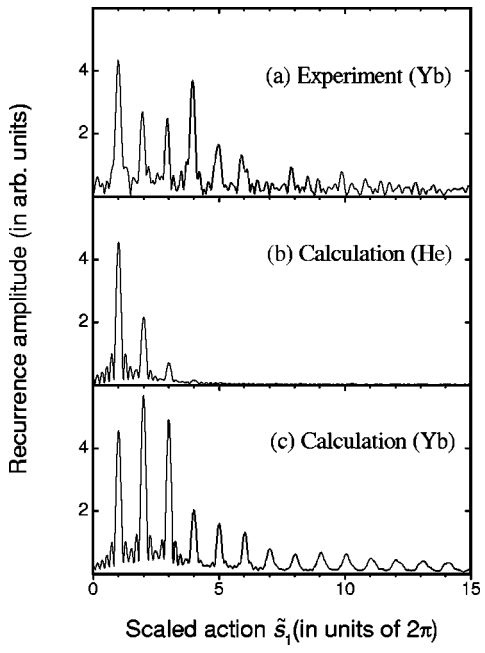

FIG. 2. (a) Experimental recurrence spectrum of Yb DRS with averaged $\bar{\varepsilon}=-0.539$ (or $\overline{N^*/n^*}=0.560$), obtained from Fourier transform of Fig. 1. Calculated recurrence spectra (b) He and (c) Yb.

By comparing Figs. 2(a) and 2(b), it can been seen that the measured peaks in Yb lie at the same actions as calculated peaks in He but, in contrast to the helium spectrum, several peaks have large amplitudes. In order to explain the origin of these peaks, we focus on the influence of the core. Classically, the core potential induces a precession of the electron orbit. This effect is incorporated into our calculation using the classical precession angle of a Kepler orbit $\Theta_L = 2\pi d\mu_L/dL$ [5] (we use a polynomial function for the quantum defect μ_L, obtained from a fit to published data [13,14]). The calculated spectrum for Yb is shown in Fig. 2(c). Comparison between Figs. 2(b) and 2(c) shows the dramatic consequences of the core on DRS. Two effects dominate. First, due to precession, new closed orbits develop in Yb, which are responsible for the peaks with $\tilde{S}_1 \gtrsim 2$ in Fig. 2(c). Second, while the recurrence of the primitive orbits is unchanged, the precession completely destroys all the repetitions. [The small He peaks with $\tilde{S}_1 \gtrsim 2$ in Fig. 2(b) disappear

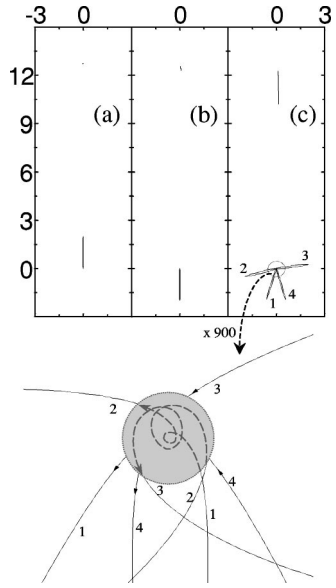

FIG. 3. Calculated classical trajectories of some closed orbits of the inner electron in the excitation of Yb DRS. (a) Primitive Zee closed orbit ($\tilde{s}_1 \simeq 1$); (b) primitive eZe closed orbit ($\tilde{s}_1 \simeq 1$); (c) a closed orbit with $\tilde{s}_1 \simeq 4$. The Yb^{2+} core is at the origin, and is denoted with a shadowed circle in the magnified region. The numbers in (c) denote the precession sequence of the orbit. Note that the outer electron (at top of each figure, close to its outer turning point) moves much slower than the inner electron (moving around the core).

completely in the case of Yb while the first peak is unchanged.]

The classical trajectories of some closed orbits for Yb are shown in Fig. 3 revealing an understanding of the shape of orbits in DRS. The core is located at the origin and the outer electron is at the top of each panel. The first (second) panel shows the Zee (eZe) primitive orbit. The third panel shows a closed classical orbit with four lobes. Lobe orientation relative to the outer electron is sensitive to electron correlation via angular momentum exchange between the two electrons. The magnified region details the trajectories near the core. Recurrence peaks are dominated by orbits where the position of the outer electron is close to its outer turning point.

A connection has been found between our calculated Zee closed orbits and frozen-planet orbits [7]. In He we find that for long times (large actions) about 2% of all orbits launched are still bound and have Zee character. These orbits exhibit the regular pattern of frozen-planet orbits. No such orbits survive in Yb. This suggests that orbit precession destroys the possibility of frozen-planet orbits, and that He is unique in its ability to support these orbits.

A recent paper [15] on the double-excitation spectrum of He provides new insight into the transition towards quantum chaos. Our work provides stimulus for investigation of the role of electron correlation at the classical-quantum interface.

In conclusion, we have applied closed-orbit theory to analyze the photoabsorption dynamics of doubly excited states. To the best of our knowledge, this is the first application of closed-orbit theory and recurrence spectroscopy to systems in which electron correlation is present. We have found that classical closed orbits exist and have studied the influence of long-range electron correlation on these orbits. We have found that both electronic correlation and quantum defects play important and different roles in the excitation. Our results show that electronic correlation destroys many but not all of the inner-electron repetition orbits, whereas precession induced by core scattering completely destroys all repetitions. As a result, in core-less helium, large action frozen-planet orbits survive but not in ytterbium. Also, new action orbits are created in Yb due to core scattering that, at certain times, kicks the electron into an orbit that has a closed returning trajectory in the presence of the dielectronic interaction. Our model captures the main features of the experimental Yb DRS recurrence spectrum and demonstrates that the approach can serve as a way to characterize quantitatively measurable properties of two-electron dynamics, and, in principle, could be extended to more complex, strongly correlated systems.

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