Photoabsorption of Nonhydrogenic Rydberg Atoms in a Magnetic Field: Effects of Core-Scattered Classical Orbits

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The physics of multielectron atoms in strong external fields is substantially more complex than that of hydrogen. In particular, puzzling new resonances were recently observed in experimental photoabsorption spectra of diamagnetic helium and in quantum mechanical calculations of diamagnetic Rydberg atoms. We compute spectra in the framework of periodic-orbit theory and show that the new resonances can be quantitatively explained by different classes of core-scattered classical orbits.

Hydrogen and nonhydrogenic atoms are real physical systems whose behavior in external fields belongs to the fundamental problems of atomic physics. The one-electron system hydrogen, which has recently been scrutinized successfully with classical, semiclassical, and quantum mechanical methods [1], is most easily accessible for theoretical treatment. In a major advance, it was proved that essential structures in quantum mechanical [2] as well as in experimental [3] photoabsorption spectra can be reproduced and interpreted semiclassically using closed classical trajectories [4,5]. Our Letter contains the first successful computation and interpretation of a multielectron spectrum in terms of periodic orbits.

The theoretical description of nonhydrogenic atoms with more than one electron in external fields is still in its infancy. Employing an *R*-matrix technique, Monteiro and Wunner [6] calculated quantum mechanical recurrence spectra of nonhydrogenic atoms and discovered novel resonance structures at positions where no hydrogenic closed orbits exist, i.e., structures which cannot be explained by hydrogenic orbits. These observations have been experimentally verified by Hogervorst and co-workers in most recent measurements of helium atoms in magnetic fields [7], and similar structures have also been discovered by Kleppner and co-workers in experimental Stark spectra of lithium [8]. Thus the search of a physical explanation of these structures means a new challenge with regard to the further development and application of semiclassical theories and methods.

In their pioneering work on a semiclassical description of nonhydrogenic atoms in external fields, Gao, Delos, and Baruch [9] incorporated quantum defects of the ionic core in the closed-orbit sum of *hydrogenic* orbits, but considered the effects of nonhydrogenic core scattering inessential. Thus core effects result only in slight shifts of resonance positions in the recurrence spectra and lead to a spectrum qualitatively different from the observed one.

Employing a short-range core potential, Dando *et al.* [10] performed classical calculations to interpret the phase-space distributions and spectral properties [11] of nonhydrogenic atoms in magnetic fields. From their analysis

they concluded that it may be necessary to consider the core scattering of semiclassical waves in the calculation of photoabsorption cross sections to explain the observed new structures in nonhydrogenic atoms in external fields. Nevertheless, it is believed uncertain [8] or problematic [10] in how far this approach can be applied to quantitatively compute the recurrence spectra.

The decisive new starting point of our treatment of nonhydrogenic atoms in the framework of semiclassical closed-orbit theory is the modeling of the ionic core by means of a suitable core potential, and the consideration of the effect of this model potential on the classical motion of the highly excited electron. The core potential obeys the following two conditions natural to general Rydberg atoms: $V(r) \xrightarrow{r \to 0} -Z/r$, with Z the nuclear charge, and $V(r) \xrightarrow{r \to \infty} -1/r$. We adopt the following dependence on r (cf. [10]):

$$V(r) = V_{\text{Coulomb}}(r) + V_{\text{core}}(r)$$
$$= -\frac{1}{r} - \frac{Z-1}{r}\left(1 + \frac{r}{a}\right)e^{-r/a}, \qquad (1)$$

where a is a free length parameter determining the range of the core. The simple ansatz of Eq. (1) is justified by our intention not to describe one particular atom in best accuracy but to study the general effects of an additional short-range potential on the photoabsorption spectra. The core potential leads to a quantum defect μ_ℓ, or for energies $E > 0$, to a phase shift δ_ℓ. At the ionization threshold, these two quantities are connected by Seaton's theorem $\delta_\ell = \pi\mu_\ell$. The quantum defects depend in general on Z, a, and the angular momentum ℓ, but decrease rapidly with increasing ℓ.

If we employ this model potential for the ionic core, the Hamiltonian of nonhydrogenic atoms reads [in atomic units, $\gamma = B/(2.35 \times 10^5 \text{ T})$]

$$H = \tfrac{1}{2}\mathbf{p}^2 + \tfrac{1}{2}\gamma L_z + \tfrac{1}{8}\gamma^2\varrho^2 + V(r). \qquad (2)$$

A special feature of the hydrogenic system is its classical scaling property [1]. After replacing $\mathbf{r} \to \gamma^{2/3}\mathbf{r}$ and $\mathbf{p} \to \gamma^{-1/3}\mathbf{p}$ the classical dynamics does not depend on the energy and field strength independently, but solely on the

scaled energy $\tilde{E} = E\gamma^{-2/3}$. The scaling property does not hold in the core region of nonhydrogenic atoms, but since this area is very small compared with the size of highly excited Rydberg states, it is appropriate to apply the scaling relations to nonhydrogenic systems as well.

The Hamiltonian of Eq. (2) served us as a basis for extensive classical trajectory calculations. We solved the equations of motion numerically and searched for all closed orbits starting at and returning to the nucleus. The effect of the additional core potential is the creation of new orbits, mainly through saddle-node and period-doubling bifurcation. The increase of the number of closed orbits is immense, e.g., for quantum defect $\mu_1 = 0.5$ and $\tilde{E} = -0.3$ we found about 2600 closed orbits with scaled action $\tilde{S} = S\gamma^{1/3} < 4.8$ compared with 35 orbits in hydrogen, and in helium at $\tilde{E} = -0.7$ about 13 000 with $\tilde{S} < 12$ compared with 42 in hydrogen.

The creation of new closed orbits can be understood as follows: A trajectory starting in almost the same initial direction as a hydrogenic closed orbit leaves the nucleus radially to evolve in the vicinity of the hydrogenic trajectory. As the electron returns to the nucleus it is scattered by the atomic core potential in an arbitrary direction which may be close to a starting angle of another hydrogenic closed orbit. Such a core scattering may be repeated several times until finally the electron exactly returns to the origin. Thus the nonhydrogenic closed orbits appear to be composed of two or more hydrogenic orbits. Upon closer examination, it turns out that one can group them together into families which consist of structurally similar orbits, if one does not consider in detail small differences in the region of the core scattering. It is natural to introduce the following notation for a family of similar orbits: we write $h_1 \oplus h_2 \oplus h_3 \oplus \cdots$, where h_i denotes the name of a primitive hydrogenic orbit and $\oplus$ indicates the scattering from one hydrogenic orbit into another by the core potential. For the hydrogenic orbits, h_i, we use the same notation as in [3]: R_μ^ν stands for types of orbits bifurcated from the motion perpendicular to the field axis, and V_μ^ν denotes the family of orbits bifurcated from the motion parallel to the field.

The recurrence time and action of the composed nonhydrogenic orbits can be approximated by the sum of the constituent hydrogenic ones, i.e., they are arranged in groups at recurrence positions where a hydrogenic orbit does not necessarily exist. In this way the new resonance structures which have recently been discovered in nonhydrogenic atoms [6–8] can be interpreted *qualitatively* by clusters of core-scattered closed classical orbits.

The tool for a *quantitative* semiclassical calculation of recurrence spectra is provided by closed-orbit theory, which has been originally developed for the hydrogen atom [4,12] and has been extended to quantum defects of a nonhydrogenic core by Gao, Delos, and Baruch [9]. Briefly, the absorbed photon excites an outgoing spherical Coulomb wave, with phase shifts given by the quantum defects of the ionic core. This wave is propagated semiclassically via classical trajectories obeying the nonhydro-

genic Hamiltonian dynamics of Eq. (2). If a trajectory returns close to the nucleus the core scattering is considered classically, i.e., by the core potential of Eq. (1). On the final return to the nucleus the semiclassical wave is matched to an incoming quantum mechanical Coulomb wave, and, loosely speaking, the interference between the outgoing and incoming waves produces modulations in the photoabsorption cross section. Details of the theoretical treatment will be published elsewhere [13].

The final result for the oscillator strength $f(E)$ in terms of closed orbits is the sum of one part f^{o}, which produces the same continuous background as in the field-free case, and an oscillatory part f^{osc},

$$
\begin{aligned}
f(E) &= f^{\mathrm{o}} + f^{\mathrm{osc}} \\
&= f^{\mathrm{o}} + 2(E - E_i)\,\mathrm{Im}\left\{\sum_{\text{closed orbits } k} A_k e^{i(S_k - \pi\alpha_k/2 + \pi/4)}\right\},
\end{aligned}
\tag{3}
$$

with E_i the energy of the initial state, S_k the classical action, and α_k the Maslov index of closed orbit k. The amplitudes A_k depend on the dipole transition operator, the starting and returning angles of the orbits, their stability, and the quantum defects of the ionic core. The contribution of a single core-scattered nonhydrogenic orbit to the closed-orbit sum (3) is usually small but this is partly compensated by constructive interference of a high number of similarly shaped orbits [13].

To compare quantum spectra of atoms in external fields with the corresponding classical dynamics it is most appropriate to apply the constant scaled energy technique [3,5]. Recurrence spectra, i.e., Fourier transforms of photoabsorption spectra, at constant scaled energy $\tilde{E} = E\gamma^{-2/3}$ taken linear in $\gamma^{-1/3}$ show detailed structures of resonances which can be interpreted semiclassically by closed orbits starting at and returning to the nucleus. In Figs. 1 and 2 we compare recurrence spectra at scaled energy $\tilde{E} = -0.3$ obtained from Ref. [6] with our results from semiclassical closed-orbit theory. Dipole transitions from an initial s state to final states with magnetic quantum number $m = 0$ are considered. The spectra of Ref. [6] were computed via an R-matrix approach, in Fig. 1(b) for vanishing quantum defects $\mu_\ell = 0$, i.e., hydrogen, and in Fig. 2(b) for a nonvanishing quantum defect $\mu_1 = 0.5$. In this spectrum pronounced nonhydrogenic resonances were discovered at $\tilde{S} \approx 3.3$ and, labeled Y, at $\tilde{S} \approx 4.4$. No closed orbits with these classical actions exist in hydrogen. We are now able to calculate and to explain these multielectron structures via core-scattered classical orbits. The hydrogenic recurrence spectrum in Fig. 1(a) is computed semiclassically with vanishing core potential. Some resonances are labeled by their corresponding closed orbits. For the nonhydrogenic atom [Fig. 2(a)] the nuclear charge Z and core size a in Eq. (1) were adjusted to achieve a quantum defect $\mu_1 \approx 0.5$ in the Fourier transformed range of magnetic field strength. As the number of closed orbits proliferates exponentially with increasing length, the classical calcu-
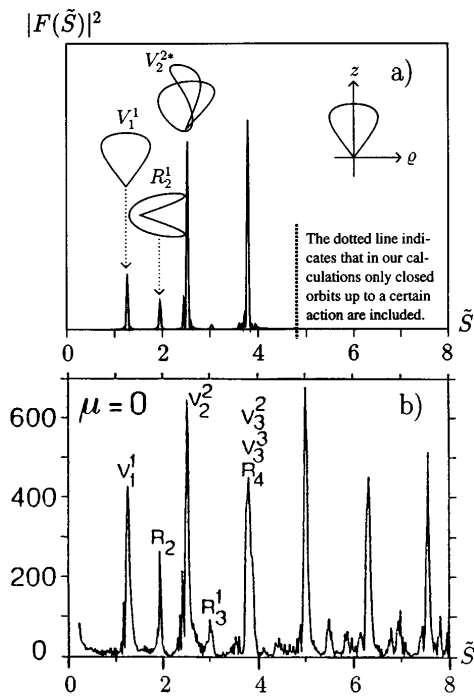
$|F(\tilde{S})|^2$

FIG. 1.　Square of Fourier transforms of $m^\pi = 0^-$ photoabsorption spectra at scaled energy $\tilde{E} = -0.3$.　(a) Hydrogenic spectrum calculated using closed-orbit theory.　(b) Hydrogenic spectrum calculated via an R-matrix approach [6].　An identification of peaks with periodic orbits has been performed using the nomenclature of Ref. [3].
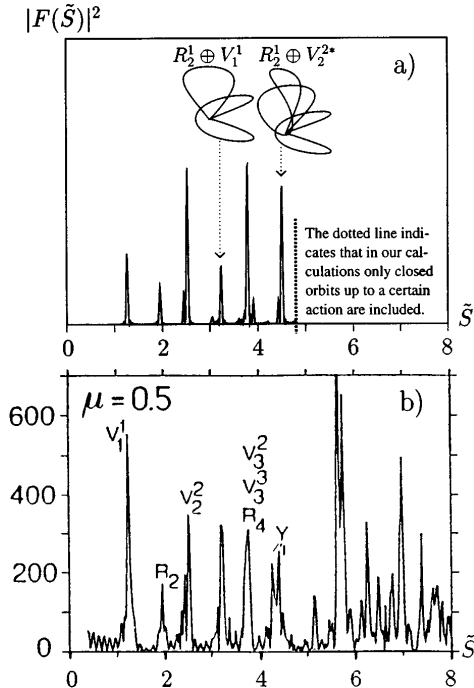
lations were restricted to $\tilde{S} < 4.8$.　In addition to the hydrogenic resonances, pronounced novel peaks are clearly visible.　The nonhydrogenic resonance at $\tilde{S} = 3.3$ belongs to a family of core-scattered orbits $R_2^1 \oplus V_1^1$.　The novel structure at $\tilde{S} \approx 4.4$ is generated by several clusters of close-by lying core-scattered orbits, the strongest contributions to the recurrence strength result from the families $R_3^1 \oplus V_1^1$ at $\tilde{S} \approx 4.32$, $R_2^1 \oplus V_2^1$ at $\tilde{S} \approx 4.45$, and $R_2^1 \oplus V_2^{2*}$ at $\tilde{S} \approx 4.51$.　For illustration two examples of core-scattered orbits are presented in Fig. 2(a).

The method can also be applied to explain recent experiments on helium in magnetic fields [7].　Hogervorst and co-workers obtained scaled energy spectra at $\tilde{E} = -0.7$ by laser excitation of metastable $1s2s^3 S_1$ atoms.　The Fourier transformed experimental recurrence spectrum is shown in Fig. 3(b).　This spectrum was compared in Ref. [7] with a numerical calculation using an R-matrix approach [Fig. 3(c)].　Both spectra clearly exhibit nonhydrogenic resonance structures marked by arrows.　For comparison numerical results for hydrogen are given in Fig. 4.　To explain the nonhydrogenic structures in helium via core-scattered classical orbits we took the nuclear charge $Z = 2.0$ in the core potential (1) and adjusted the core size a to achieve a quantum defect μ_1 close to the experimental value $\mu_1 = 0.0684$.　The semiclassical recurrence spectrum is presented in Fig. 3(a).　The novel resonances result from single core-scattered closed orbits $R_\mu^1 \oplus V_\nu^1$ with μ

FIG. 2.　Square of Fourier transforms of $m^\pi = 0^-$ photoabsorption spectra at scaled energy $\tilde{E} = -0.3$ and quantum defect $\mu_1 = -0.5$.　(a) Nonhydrogenic spectrum calculated using closed-orbit theory.　(b) Nonhydrogenic spectrum calculated via an R-matrix approach [6].　In both spectra the same new resonances appear, which can now be identified with core-scattered periodic orbits.
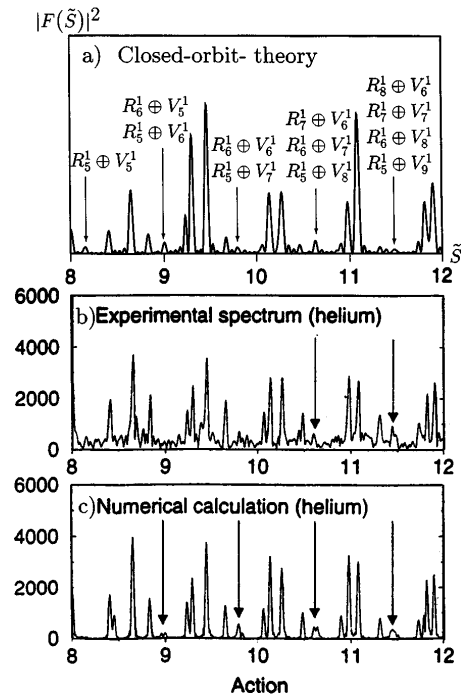

FIG. 3.　Square of Fourier transforms of $m^\pi = 0^-$ photoabsorption spectra at scaled energy $\tilde{E} = -0.7$ and a quantum defect μ_1 corresponding to helium.　(a) Helium spectrum calculated using closed-orbit theory.　(b) Experimental spectrum of helium [7].　(c) Spectrum calculated via an R-matrix approach [7].　Obviously, the new resonances in helium can be explained in terms of core-scattered classical closed orbits.
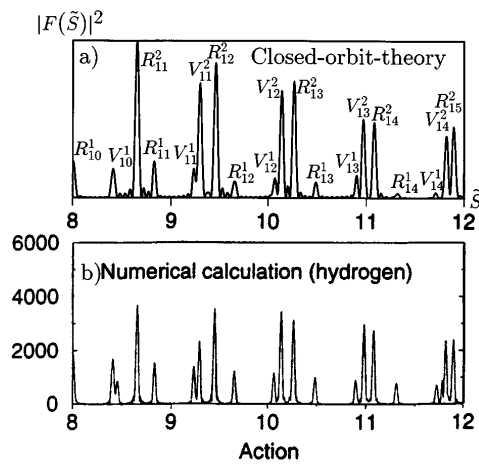
FIG. 4. Square of Fourier transforms of $m^\pi = 0^-$ photoabsorption spectra at scaled energy $\tilde{E} = -0.7$. (a) Hydrogenic spectrum calculated using closed-orbit theory. (b) Hydrogenic spectrum calculated via an R-matrix approach. (From Ref. [7].) An identification of peaks with periodic orbits using the nomenclature of Ref. [3] is also given.

and ν between 5 and 9, which are indicated in Fig. 3(a), as well as from multiple core-scattered orbits. Although the orbits of the latter families have low amplitude, they contribute significantly because their number is enormous. It should be remarked that the orbits R_5^1 and V_5^1, appearing as parts of composite nonhydrogenic orbits (e.g., at $\tilde{S} = 8.2$), do not exist as single closed orbits because they are born in hydrogen at scaled energies slightly above $\tilde{E} = -0.7$.

In summary, we have calculated Fourier transformed spectra at constant scaled energy of general Rydberg atoms in magnetic fields using a closed-orbit theory extended to arbitrary quantum defects, and taking into account an additional core potential. We compared our spectra with experiments and quantum calculations and could successfully interpret previously puzzling deviations from hydrogen to nonhydrogenic atoms. Our method should also allow the explanation of nonhydrogenic features in Stark spectra of lithium [8] or in rubidium atoms in crossed magnetic and electric fields [14], to cite two examples, and thus aid in gaining a deeper understanding of the behavior of multielectron Rydberg atoms in external fields.

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