

Semiclassical quantization with bifurcating orbits

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Bifurcations of classical orbits introduce divergences into semiclassical spectra which have to be smoothed with the help of uniform approximations. We develop a technique to extract individual energy levels from semiclassical spectra involving uniform approximations. As a prototype example, the method is shown to yield excellent results for photoabsorption spectra for the hydrogen atom in an electric field in a spectral range where the abundance of bifurcations would render the standard closed-orbit formula without uniform approximations useless. Our method immediately applies to semiclassical trace formulas as well as the closed-orbit theory and offers a general technique for the semiclassical quantization of arbitrary systems.

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The correspondence between atomic spectra and classical orbits has been of fundamental interest and importance since the early days of quantum mechanics. The “old” quantum theory suffered from the severe drawbacks that the Bohr-Sommerfeld quantization rules can only be applied to integrable systems, and, for atomic systems, the Heisenberg principle for matrix elements is silent about transition amplitudes between low-lying and highly excited states. An important step towards a deeper understanding of the intimate connection between classical orbits and the quantum spectra was achieved by the development of the periodic orbit theory [1] and, as a variant for the photoexcitation of atomic systems, closed orbit theory [2,3]. In these theories, the density of states or photoabsorption spectra are given as the sum of two terms, one a smoothly varying part (as a function of energy) and the other a superposition of sinusoidal modulations. The frequencies, amplitudes, and phases of the modulations are directly given in terms of classical parameters of the orbits.

The closed-orbit theory has proven a powerful tool for the semiclassical interpretation of quantum spectra of, e.g., atoms in external magnetic and electric fields by explaining the peaks in the Fourier-transform recurrence spectra—qualitatively and even quantitatively—in terms of the closed orbits of the underlying classical system [4–6]. However, more than a decade after the development of the closed-orbit theory, the inverse procedure, i.e., the semiclassical calculation of the eigenenergies and transition amplitudes of individual states is still an unsolved problem. The reasons are twofold: First, both the closed-orbit and periodic-orbit theories suffer from fundamental convergence problems of the infinite orbit sums. Second, in generic systems the orbits undergo bifurcations when the energy is varied, and the semiclassical theories for isolated orbits exhibit unphysical divergences at the bifurcation points. Both problems have been addressed separately: First, the harmonic inversion technique was introduced as a method for semiclassical quantization [7,8], which allows one to overcome the convergence problems of the closed orbit sum and to extract high-resolution spectra from a finite set of classical orbits. Second, in the vicinity of bifurcations, the semiclassical approximation for isolated orbits was replaced with a uniform approximation describing all orbits involved in a bifurcation collectively [9,10].

In this paper both ideas are combined, i.e., we use both uniform approximations and harmonic inversion techniques for the semiclassical calculation of high-resolution spectra. In the presence of uniform approximations, the classical scaling laws that have been essential to all previous applications of the harmonic inversion technique [8] are no longer valid. Therefore, the harmonic inversion method must be generalized to handle the nonscaling functional form of uniform semiclassical approximations. It then gains a degree of flexibility in the quantization of arbitrary systems, which no other semiclassical quantization scheme has been able to reach to date.

The proposed method will be demonstrated by way of an example of the hydrogen atom in an electric field. As is well known, the classical dynamics of this system is integrable, which means that semiclassical energy eigenvalues can be calculated with the help of the Einstein-Brillouin-Keller torus quantization rules [11,12]. However, when the closed-orbit theory is applied, the hydrogen atom in an electric field exhibits properties typical of mixed regular-chaotic systems as, e.g., Rydberg atoms in a magnetic field or Hénon-Heiles-type systems. In particular, the closed orbits starting at and returning to the nucleus undergo bifurcations as the energy is varied. Contrary to the torus quantization, the method introduced in this paper is not restricted to the Stark effect, but can be applied to a large variety of systems with chaotic or mixed regular-chaotic classical dynamics. Furthermore, it can be used in connection with the periodic-orbit theory [1] as well as the closed-orbit theory.

The classical dynamics of the Stark system has already been discussed in detail [11]. For any energy, the electron can go “uphill” against the direction of the electric field until the external field and the Coulomb field make it return to the nucleus. Alternatively, the electron can leave the nucleus in the “downhill” direction of the external field. The downhill orbit is closed only for energies below the Stark saddle-point energy, $E_S = -2F^{1/2}$, otherwise the electron will cross the Stark saddle and escape to infinity. In addition to these axial closed orbits, there are nonaxial orbits returning to the nucleus after k oscillations in the downhill direction and $l > k$ oscillations in the uphill direction. Each of these orbits is generated in a bifurcation off the downhill orbit at a critical

energy E_{gen} and destroyed in a collision with the uphill orbit at $E_{\text{dest}} > E_{\text{gen}}$. Outside this energy range, they exist as “ghost” orbits in the analytic continuation of the real phase space to complex numbers.

The closed-orbit theory associates modulations observed in the quantum photoabsorption spectra of Rydberg atoms in external fields with the classical closed orbits. The quantum response function

$$g(E) = \sum_n \frac{|\langle i|D|n\rangle|^2}{E - E_n + i\epsilon} = \langle i|D G_E^+ D|i\rangle, \quad (1)$$

where $|i\rangle$ is the initial state, D is the dipole operator, and G_E^+ is the retarded Green’s function, is given as a smooth background plus an oscillatory closed-orbit sum [2,3,13]

$$g^{\text{osc}}(E) = \sum_{\text{co}} \mathcal{A}_{\text{co}}(E) e^{iS_{\text{co}}(E)}, \quad (2)$$

where S_{co} is the action of a closed orbit (co) and \mathcal{A}_{co} a recurrence amplitude calculated from the monodromy matrix of the orbit and its initial and final directions with respect to the electric field. It includes a complex phase given by the Maslov index. In the following, we are using atomic units, with $\hbar = 1$ and $F_0 = 5.14 \times 10^9$ V/cm being the unit of the electric field strength.

The most convincing semiclassical interpretation of quantum spectra can be obtained by means of “scaled energy spectroscopy:” By rescaling the classical quantities with suitable powers of, e.g., the electric field strength F , the classical dynamics can be shown not to depend on the energy E and the field strength F separately, but only on the scaled energy $\tilde{E} = EF^{-1/2}$. When recording quantum states at a fixed scaled energy \tilde{E} as a function of the scaling parameter $w = F^{-1/4}$, each isolated closed orbit contributes a sinusoidal modulation to the sum (2), which can be extracted by a Fourier transform of the quantum spectrum. Experimental scaled energy spectra of atoms in electric fields have been analyzed in this way [5,6]. The analysis reveals strong evidence for closed-orbit bifurcations.

The simple semiclassical approximation embodied in the closed-orbit formula (2) fails close to a bifurcation of closed orbits, resulting in the divergence of the recurrence amplitudes. To overcome this difficulty, the closed-orbit terms for isolated orbits in Eq. (2) must be replaced with a uniform approximation describing all orbits involved in a bifurcation collectively. A uniform approximation suitable for regularizing the bifurcation of a nonaxial orbit off either the downhill or the uphill orbit was derived by Gao and Delos [10] as well as Shaw and Robicheaux [14]. We will use a slightly modified version of their result that gives the collective contribution of the axial and nonaxial orbits participating in a bifurcation in terms of their actions S_{ax} and S_{non} and recurrence amplitudes \mathcal{A}_{ax} and \mathcal{A}_{non} as

$$\Psi(E) = \left[\frac{\mathcal{A}_{\text{non}}}{(1+i)} I + \frac{1}{a} \left(a\mathcal{A}_{\text{ax}} + \frac{1+i}{\sqrt{2\pi}} \mathcal{A}_{\text{non}} \right) \right] e^{iS_{\text{ax}}}, \quad (3)$$

where I is given in terms of the standard Fresnel integrals $C(x)$ and $S(x)$ [15],

$$I = e^{-ia^2/4} \left[\frac{1+i}{2} - C\left(\frac{-a}{\sqrt{2\pi}}\right) - iS\left(\frac{-a}{\sqrt{2\pi}}\right) \right], \quad (4)$$

and

$$a = \pm 2\sqrt{S_{\text{ax}} - S_{\text{non}}}. \quad (5)$$

The negative sign for a has to be chosen if the nonaxial orbit is a complex ghost orbit.

The high-resolution quantization by harmonic inversion [7,8] is based on the observation that by equating the quantum recurrence function (1) to its semiclassical approximation (2)—the smooth part can be neglected—and taking the Fourier transform, we obtain

$$-i \sum_n d_n e^{-iE_n t} = C(t) \quad (6)$$

with $d_n = |\langle i|D|n\rangle|^2$ and

$$C(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dE \sum_{\text{co}} \mathcal{A}_{\text{co}}(E) e^{iS_{\text{co}}(E)} e^{-iEt}. \quad (7)$$

The quantization problem has thus been recast as the problem of extracting the frequencies E_n and amplitudes d_n from a given time signal $C(t)$ of the form (6), provided the signal (7) can be calculated. In the case of a scaling system, the signal is given as a sum of δ functions.

While the uniform approximation (3) successfully smoothes the divergences in Eq. (2), it spoils the classical scaling properties [14]. Therefore, the Fourier transform of a spectrum including uniform approximations cannot be evaluated in terms of δ functions. In fact, for nonscaling systems, there seems to be no way at all to compute the integral (7), because, apart from the fact that the classical quantities can always be calculated in a finite-energy interval only, the integral can, in general, not even be expected to converge. Therefore, neither by analytical nor by numerical means will one be able to compute a useful semiclassical signal from (7). The inclusion of uniform approximations in semiclassical quantization is thus a nontrivial and challenging task.

To solve the problem, we resort to the observation made in Ref. [16] that a band-limited signal, which only contains the spectral information describing the quantum system in a finite-energy interval $[E_{\text{min}}, E_{\text{max}}]$, can be obtained by restricting the energy integral in Eq. (7) to this window. The resulting signal

$$C^{\text{bl}}(t) = \frac{1}{2\pi} \int_{E_{\text{min}}}^{E_{\text{max}}} g^{\text{osc}}(E) e^{-iEt} dE, \quad (8)$$

where $g^{\text{osc}}(E)$ is in general a nonscaling function that includes both contributions of isolated closed orbits and uniform approximations, can be evaluated numerically by standard methods. As it contains only a small number of frequencies in the interval chosen, it can be processed in a

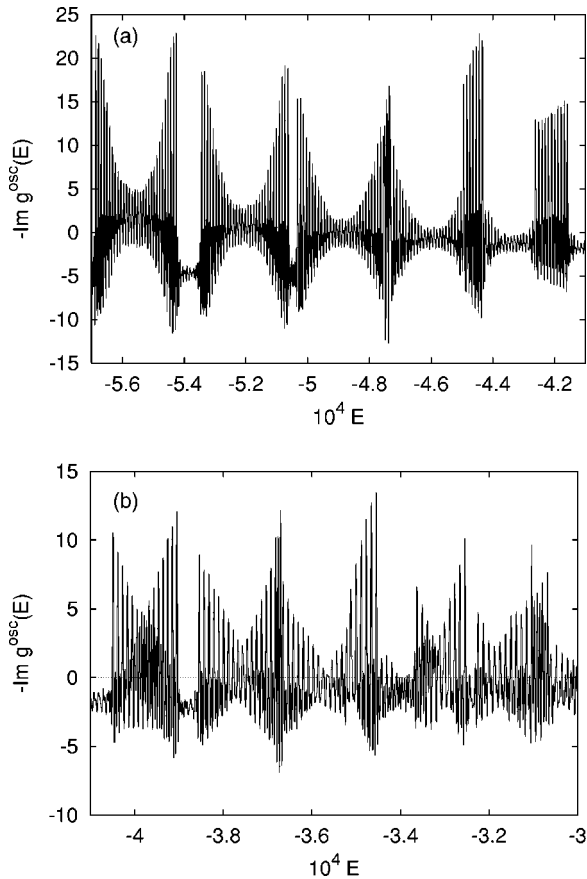


FIG. 1. Low-resolution semiclassical photoabsorption spectrum for the hydrogen atom in an electric field $F = 51.4$ V/cm with initial state $|1s0\rangle$ and light polarized along the electric field axis. The truncation time is $T_{\max} = 15 \times 10^6$.

numerically stable way by conventional high-resolution methods such as linear prediction or Padé approximants. Contrary to previous methods for scaling systems, all of which contained the analytic evaluation of an integral, the numerical integration imposes no restrictions on the semiclassical response function occurring in the integrand. Notice that our method cannot be applied in connection with the original filter diagonalization algorithm [8]. It is only the separation between a low-resolution frequency filtering stage and a high-resolution harmonic inversion stage introduced in Ref. [16] that allows for the present generalization to arbitrary nonscaling semiclassical signals.

To demonstrate our method we investigate Stark spectra of the hydrogen atom for transitions from the ground state $|1s0\rangle$ to highly excited Rydberg states with light polarized parallel to the electric-field axis. The external field strength is $F = 10^{-8}$ a.u. $\hat{=} 51.4$ V/cm. The high-resolution Stark spectrum is obtained in two steps.

First, a low-resolution semiclassical spectrum is obtained by truncating the closed-orbit sum (2) at a maximal period T_{\max} . The cutoff value for the periods should not be chosen smaller than the signal length of the band-limited signal (8) used for the harmonic inversion in the second step of the procedure. The low-resolution spectrum calculated with a truncation time of $T_{\max} = 15 \times 10^6$ is presented in Fig. 1. In

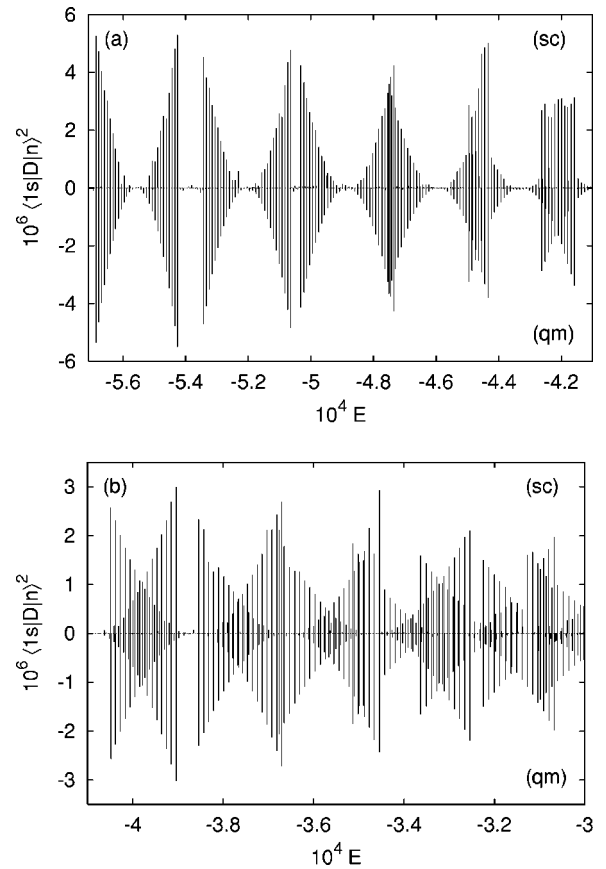


FIG. 2. High-resolution semiclassical (upper part) and quantum (lower part, inverted) photoabsorption spectrum for the hydrogen atom in an electric field $F = 51.4$ V/cm with initial state $|1s0\rangle$ and light polarized along the electric field axis. The truncation time for the semiclassical spectrum is $T_{\max} = 40 \times 10^6$.

the lower-energy range shown in Fig. 1(a), individual non-overlapping n manifolds can be observed. (We have $n = 30$ at $E \approx -5.56 \times 10^{-4}$.) In this region, the signal is sufficiently long to resolve individual spectral lines, although their precise determination from the plots remains difficult. In the higher-energy range shown in Fig. 1(b), two, three, or even four different n manifolds overlap, leading to a drastically increased spectral density. In this region the semiclassical signal is evidently too short to discriminate individual lines. It is important to note that the possibility of computing the low-resolution spectrum depends critically on the use of uniform approximations. If it was calculated from isolated-orbit contributions only, a dense sequence of bifurcation-induced divergences would cover even the large-scale structure of the spectra. With the low-resolution semiclassical spectrum (Fig. 1) at hand, the band-limited time signal (8) is now obtained by a numerical Fourier transform of $g^{\text{osc}}(E)$. In that calculation we used the signal length $T_{\max} = 40 \times 10^6$ in order to resolve individual levels in the region of overlapping n manifolds.

In the second step, the high-resolution semiclassical spectrum is finally obtained by harmonic inversion of the band-limited time signal (8). In Fig. 2 the semiclassical spectrum is compared to the exact quantum spectrum. The overall

agreement between the semiclassical and the quantum spectrum is excellent, although for a few levels the comparison reveals discrepancies between the semiclassical and the quantum matrix elements. Note, in particular, the region of high spectral density at $E \approx 3.2 \times 10^{-4}$. In this region, groups of three nearly degenerate levels exist, some of which are well resolved semiclassically. At $E \approx -4.4 \times 10^{-4}$, even closer lines exist—they can hardly be discerned in the quantum spectrum. These lines are not resolved semiclassically. Instead, the harmonic inversion yields single lines with amplitudes equal to the sum of the two quantum amplitudes. We are confident to fully resolve even these states in the semiclassical spectrum when applying the cross-correlation technique for harmonic inversion [8].

In the future it should also be possible to extend the semiclassical spectrum to energies $E > E_S$, where the classical motion is not completely bound, and to extract the semiclassical widths of the Stark resonances. A particular challenge is posed by the region around the Stark saddle-point energy E_S . Before the downhill orbit ceases to exist at E_S , it undergoes an infinite sequence of bifurcations, giving birth to nonaxial orbits with arbitrarily high uphill repetition numbers. If subsequent bifurcations of a single orbit are too close, the uniform approximation (3) is no longer appropri-

ate. It must then be replaced with a uniform approximation describing several bifurcations collectively. The uniformization of an infinite bifurcation cascade, in particular, remains an open problem whose solution is required to semiclassically cross the saddle-point energy.

In summary, we have extended the harmonic inversion approach to semiclassical quantization to the quantization of systems without a scaling property. The generalized method allows for the inclusion of uniform approximations into the quantization procedure. We have demonstrated the effectiveness of our method by calculating a high-quality semiclassical spectrum for the hydrogen atom in an electric field in a spectral region where the semiclassical approximation without uniform approximations would be completely useless due to the abundance of bifurcations. With the modifications presented here, the technique of quantization by harmonic inversion has reached a stage where it does not impose any conditions on the classical dynamics of the system under study, except that a semiclassical approximation to the response function can be given. Besides uniform approximations, any other nonstandard semiclassical contributions such as diffractive corrections can be included. Thus, the harmonic inversion can now be regarded as a truly universal tool for the semiclassical quantization of arbitrary systems.

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