

## High Resolution Quantum Recurrence Spectra: Beyond the Uncertainty Principle

Jörg Main,<sup>1,2</sup> Vladimir A. Mandelshtam,<sup>2</sup> and Howard S. Taylor<sup>2</sup>

<sup>1</sup>*Institut für Theoretische Physik I, Ruhr-Universität Bochum, D-44780 Bochum, Germany*

<sup>2</sup>*Department of Chemistry, University of Southern California, Los Angeles, California 90089*

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Highly resolved recurrence spectra are obtained by harmonic inversion of quantum spectra of classically chaotic systems and compared in detail to the results of semiclassical *periodic orbit* and *closed orbit* theory. Our analysis is sensitive to separate orbits with nearly degenerate recurrence periods and uncovers complex (“ghost”) orbits even when they are hidden behind close-by real orbits. The method is demonstrated on an example of the hydrogen atom in external magnetic and electric fields, for both the density of states and the quantum photoabsorption cross section. [S0031-9007(97)03393-0]

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Semiclassical *periodic orbit* [1] and *closed orbit* [2,3] theory has become the key for the interpretation of quantum spectra of classically chaotic systems. The semiclassical spectra at least in low resolution are given as the sum of a smooth background and a superposition of modulations whose amplitudes, frequencies, and phases are solely determined by the closed or periodic orbits of the classical system. For the interpretation of quantum spectra in terms of classical orbits it is therefore most natural to obtain the recurrence spectrum by Fourier transforming the energy spectrum into the time domain. Each closed or periodic orbit should show up as a sharp  $\delta$ -peak at the recurrence time (period), provided first, the classical recurrence times do not change within the whole range of the spectrum and, second, the Fourier integral is calculated along an infinite energy range. The first condition can be fulfilled in systems possessing a classical scaling property in that the classical dynamics does not change if the energy and an external scaling parameter are simultaneously varied. An example is the hydrogen atom in crossed magnetic and electric fields (for reviews see [4–6]) given by the Hamiltonian [in atomic units,  $\gamma = B/(2.35 \times 10^5 \text{ T})$ ,  $f = F/(5.14 \times 10^9 \text{ V/cm})$ ]

$$H = \frac{1}{2} \mathbf{p}^2 - \frac{1}{r} + \frac{1}{2} \gamma L_z + \frac{1}{8} \gamma^2 (x^2 + y^2) + fx. \quad (1)$$

The classical dynamics does not depend on three parameters, the energy  $E$  and the two field strengths  $\gamma$  and  $f$ , but solely on the scaled energy  $\tilde{E} = E\gamma^{-2/3}$  and the scaled field strength  $\tilde{F} = f\gamma^{-4/3}$ . Applying the method of *scaled energy spectroscopy* [7,8] the quantum spectra can be created at a constant scaled energy as a function of the scaling parameter  $w \equiv \gamma^{-1/3}$ . The scaled spectra can be Fourier transformed along arbitrarily long ranges of  $w$  to generate Fourier transform recurrence spectra of in principle arbitrarily high resolution. By varying the scaled energy a direct comparison of the quantum recurrence spectra with the bifurcation diagram of the underlying classical system is possible [7,8]. However, the second condition is never fulfilled in practice, i.e., the

length of quantum spectra is always restricted either by experimental limitations or, in theoretical calculations, by the growing dimension of the Hamilton matrix which has to be diagonalized numerically. The given length of a quantum spectrum determines the resolution of the quantum recurrence spectrum due to the uncertainty principle,  $\Delta E \Delta T \sim \hbar$ , when the conventional Fourier transform is used. Only those closed or periodic orbits can be clearly identified quantum mechanically which appear as isolated nonoverlapping peaks in the quantum recurrence spectra. This is especially not the case for orbits which undergo bifurcations at energies close to the bifurcation point.

In this Letter we present a method to calculate *high resolution recurrence spectra* from experimental or theoretical quantum spectra of *finite length* thereby circumventing the restrictions of the uncertainty principle to the resolution of finite range Fourier transforms. It allows one, e.g., to identify real orbits with nearly degenerate periods and to detect complex “ghost” orbits which can be of particular importance in the vicinity of bifurcations [9,10]. According to *periodic orbit* theory [1] the semiclassical density of states can be written as the sum of a smooth background  $\varrho_0(w)$  and oscillatory modulations induced by the periodic orbits,

$$\varrho(w) = \varrho_0(w) + \text{Im} \sum_k A_k e^{-2\pi i \tilde{S}_k w}. \quad (2)$$

The amplitude  $A_k$  and scaled action  $\tilde{S}_k = (2\pi w)^{-1} \oint_k \mathbf{p} \cdot d\mathbf{r}$  of periodic orbit  $k$  are obtained from classical calculations and are in general complex quantities. The amplitude  $A_k$  contains the phase information determined by the Maslov index of the orbit. A nonreal action  $S_k$  indicates a ghost orbit [9,10] which exists in the complex continuation of the classical phase space. A similar formula as Eq. (2) also holds in *closed orbit* theory [2,3] to calculate semiclassically the photoabsorption cross section for dipole transitions from an energetically low lying initial state. Oscillatory modulations of the cross section are induced by closed orbits of the electron starting at and returning back to the nucleus. [In closed orbit theory the oscillatory terms of

the photoabsorption cross section are proportional to  $w^\alpha$  with  $\alpha = -1/2$  for the hydrogen atom in a magnetic field and  $\alpha = -1$  for the atom in crossed magnetic and electric fields. To obtain the functional form of Eq. (2) we multiply the cross section by  $w^{-\alpha}$ .

It might appear that the problem of identifying  $A_k$  and  $\tilde{S}_k$  that contribute to the quantum spectrum  $\varrho(w)$  can be solved by Fourier transforming  $\varrho(w)$ . As was already pointed out the low resolution nature of the Fourier transform for a short signal does not permit one to resolve closely spaced peaks, neither does it allow one to distinguish between real and ghost orbits. Instead of using the standard Fourier analysis, to extract the amplitudes and actions in this Letter we propose to essentially fit a finite range of the quantum spectrum by the semiclassical expression (2) with unknown and in general complex parameters  $A_k$  and  $\tilde{S}_k$ . The problem of fitting a "signal"  $\varrho(w)$  to the functional form (2) is known as *harmonic inversion* with a large variety of applications in various fields [11]. As a numerical technique for the harmonic inversion of a signal, i.e. a quantum spectrum, we apply the method of filter diagonalization [12] which allows us to extract the frequencies (here actions  $\tilde{S}_k$ ) in any given interval of interest. The method was recently improved in that it allows one to significantly reduce the required length of the analyzed signal [13]. Operationally, one proceeds by setting up a small generalized eigenvalue problem requiring as an input only the knowledge of the signal  $\rho(\omega)$  on a finite evenly spaced grid. The complex actions  $\tilde{S}_k^{\text{qm}}$  in the chosen interval and amplitudes  $A_k^{\text{qm}}$  are obtained from the resulting eigenvalues and eigenvectors. Thus, the recurrence spectrum is effectively discretized, the number of terms being the number of eigenvalues in the spectral domain. This method is a variational one (as opposed to the Fourier transform) and therefore has practically an infinite resolution once the amount of information contained in the signal  $\varrho(w)$  is greater than the total number of unknowns  $A_k$  and  $S_k$ . The minimal length of the quantum spectra required for the convergence of individual closed or periodic orbit parameters depends linearly on the average local density of orbits at that period and not the inverse minimal spacing. As such the "uncertainty principle" being the very property of the Fourier transform is irrelevant in the present context.

In quantum calculations the bound state spectrum is given as a sum of  $\delta$  functions. In order to obtain  $\varrho(w)$  on an evenly spaced grid the spectrum is regularized by convoluting it with a narrow Gaussian function having the width  $\sigma \ll 1/\tilde{S}_{\text{max}}$ , where  $S_{\text{max}}$  is the scaled action of the longest orbit of interest. The quantum spectrum reads  $\varrho(w) = w^{-\alpha} \sum_n D_n \delta_\sigma(w - w_n)$ , where  $w_n$  are the eigenvalues of the scaling parameter  $w$  and the weights  $D_n$  are, e.g., the absolute value squared of transition matrix elements for excitations from a given initial state. The unweighted density of states is obtained with  $\alpha = 0$

$D_n = 1$ . By varying the Gaussian width  $\sigma$  we carefully checked that it was chosen sufficiently small not to effect the results of our harmonic inversion analysis.

We now demonstrate on the hydrogen atom in a magnetic field and in crossed magnetic and electric fields that physically interesting effects can be revealed in the quantum spectra by application of the harmonic inversion technique. We do not present the complete high resolution recurrence spectra here but concentrate on the physically interesting parts which cannot be resolved by conventional Fourier transform.

*Precision check of the periodic orbit theory.*—As a first example we apply the method to the density of states for the hydrogen atom in a magnetic field at scaled energy  $\tilde{E} = -0.1$ . At this energy the classical dynamics is completely chaotic and all periodic orbits are unstable. We calculated 9715 states in the region  $w < 140$  by numerical diagonalization of the Hamiltonian matrix in a complete basis set. For details of the quantum calculations see, e.g., Ref. [14]. The quantum density of states was analyzed both by conventional Fourier transform and by the high resolution method [13]. To get rid of unphysical side peaks in the conventional Fourier transform we multiply the spectra with a Gaussian window which is chosen in accordance with the total length of the quantum spectrum. A comparison between the Fourier transform, the high resolution quantum recurrence spectrum, and the semiclassical recurrence spectrum is presented in Fig. 1 around scaled action  $\tilde{S} = 2.6$ . The smooth line is the absolute value of the conventional Fourier transform. Its shape suggests the existence of at least three periodic orbits but obviously the recurrence spectrum is not completely resolved. The results of the high resolution spectral analysis are presented as sticks at the

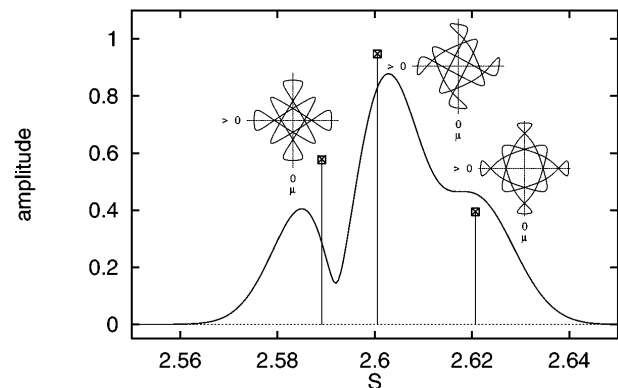


FIG. 1. Recurrence spectrum for the density of states of the hydrogen atom in a magnetic field at scaled energy  $\tilde{E} = E\gamma^{-2/3} = -0.1$ . Smooth line: Conventional Fourier transform. Solid stick spectrum and crosses: High resolution quantum recurrence spectrum. Dashed sticks (hardly visible under solid sticks) and squares: Recurrence spectrum from semiclassical *periodic orbit* theory. The recurrence peaks are identified by periodic orbits drawn in semiparabolic coordinates  $\mu = (r + z)^{1/2}$ ,  $\nu = (r - z)^{1/2}$ .

positions defined by the scaled actions  $\tilde{S}_k^{\text{qm}}$  with peak heights  $|A_k^{\text{qm}}|$ . Note that the positions of the peaks are considerably shifted with respect to the maxima of the conventional Fourier transform. To compare the quantum recurrence spectrum with Gutzwiller's periodic orbit theory we calculated the classical scaled actions  $\tilde{S}_k^{\text{cl}}$  of the periodic orbits and their semiclassical amplitudes  $|A_k^{\text{cl}}| = \tilde{S}_k^{\text{cl}} / \sqrt{|\det(M_k - I)|}$ , with  $M_k$  being the monodromy matrix of orbit  $k$ . The semiclassical results are presented as dashed sticks and squares in Fig. 1. For illustration the shapes of periodic orbits are also shown (in semiparabolic coordinates  $\mu = \sqrt{r+z}$ ,  $\nu = \sqrt{r-z}$ ). For these three orbits the agreement between the semiclassical and the high resolution quantum recurrence spectrum is nearly perfect, deviations are within the stick widths. For example, for the first peak in Fig. 1 we find  $(\tilde{S}^{\text{cl}} - \tilde{S}^{\text{qm}}) / \tilde{S}^{\text{qm}} = 8 \times 10^{-6}$  and  $(|A^{\text{qm}}| - |A^{\text{cl}}|) / |A^{\text{qm}}| = 0.0043$ . The deviation between the classical and quantum amplitudes might be related to higher order  $\hbar$  corrections of the semiclassical trace formula. In fact, Gutzwiller's trace formula is only the first term of the semiclassical series, and the high resolution recurrence spectra might be a sensitive tool to identify these corrections quantitatively in quantum spectra.

*Uncovering the "hidden" ghost orbits.*—In systems where classical orbits undergo bifurcations complex ghost orbits may be important for a complete understanding of quantum spectra, as was shown for the kicked top [9] and for the hydrogen atom in a magnetic field [10]. The contribution of a ghost orbit decreases exponentially with the energy distance from the bifurcation point and therefore ghosts can be detected in quantum spectra by conventional Fourier transform only if they are isolated from the other orbits, e.g., as a prebifurcation ghost of a saddle-node bifurcation [9]. For other types of bifurcations, e.g., for the bifurcation of the perpendicular orbit of the hydrogen atom in a magnetic-field ghost orbits are hidden behind a strong recurrence peak of a real orbit with nearly the same classical action [10]. Such hidden ghosts can now be uncovered in high resolution quantum recurrence spectra. The key point is that the harmonic inversion of quantum spectra supplies complex frequencies  $\tilde{S}_k^{\text{qm}}$  in Eq. (2) which can be interpreted as complex actions  $\tilde{S}_k^{\text{cl}}$  of ghost orbits. In Fig. 2 we have analyzed the photoabsorption spectrum of the hydrogen atom in a magnetic field at scaled energy  $\tilde{E} = -0.35$  in the region  $w < 100$  (2823 transitions from the initial state  $|2p0\rangle$  to final states with  $m^{\pi z} = 0^+$ ). The conventional Fourier transform (smooth line) has a maximum at  $\tilde{S} = 1.895$  which is roughly twice the period of the perpendicular orbit but does not give any hint on the existence of a ghost orbit. The high resolution spectral analysis uncovers one real and two complex actions  $\tilde{S}_k^{\text{qm}}$  which are compared to the classical actions  $\tilde{S}_k^{\text{cl}}$  of the perpendicular orbit and a complex ghost [see crosses and squares in Fig. 2(b), respectively]. For the frequency with the largest imaginary part no classical

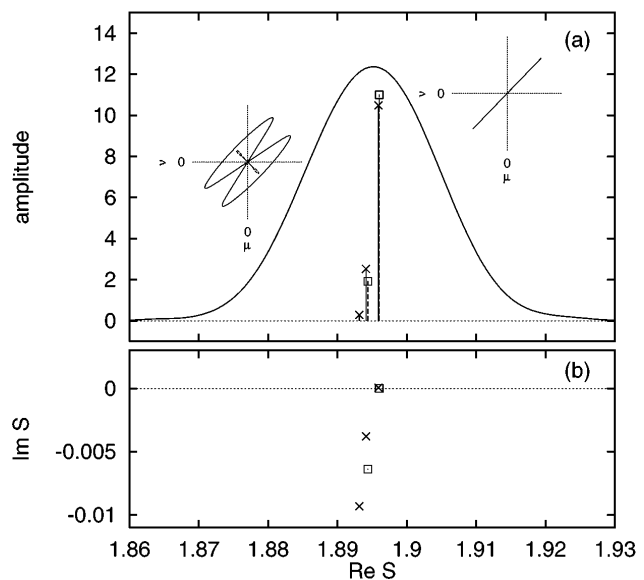


FIG. 2. (a) Recurrence spectrum for the photoabsorption cross section of the hydrogen atom in a magnetic field at scaled energy  $\tilde{E} = E\gamma^{-2/3} = -0.35$ . Transition  $|2p0\rangle \rightarrow |m^{\pi z} = 0^+\rangle$ . Smooth line: Conventional Fourier transform. Solid stick spectrum and crosses: High resolution quantum recurrence spectrum. Dashed sticks and squares: Recurrence spectrum from semiclassical *closed orbit* theory. The two strongest recurrence peaks are identified by a real and complex ghost orbit. The solid and dashed lines are the real and imaginary parts in semiparabolic coordinates  $\mu = (r+z)^{1/2}$ ,  $\nu = (r-z)^{1/2}$ . (b) Complex actions. Crosses and squares are the quantum and classical results, respectively.

analog has been found, but its amplitude [see Fig. 2(a)] is rather small. For the two stronger contributions the agreement between quantum and classical recurrence spectrum is remarkably good. The shapes of the real and complex closed orbits are presented as insets in Fig. 2(a) (in semiparabolic coordinates  $\mu, \nu$ ). The real part of the ghost orbit (solid line) is similar to the "pacman" orbit  $R_2^1$  (in the notation of [10]) which is created as a real orbit at much higher energy  $\tilde{E} = -0.317$ . This is the first observation of a hidden ghost orbit in quantum mechanical spectra.

*Breaking the cylindrical symmetry in crossed magnetic and electric fields.*—The hydrogen atom in a magnetic field has a cylindrical symmetry around the magnetic-field axis; i.e., closed and periodic orbits do not depend on the azimuthal starting angle  $\varphi$ . This symmetry is broken in crossed magnetic and electric fields. Out of a manifold of closed orbits only two closed orbits with slightly different classical actions survive in the crossed fields [15]. As a result each recurrence peak splits into two. We investigate the symmetry breaking in the crossed field atom at scaled energy  $\tilde{E} = -0.5$  and scaled field strength  $\tilde{F} = 0.02$  where we calculated the photoabsorption spectrum (transitions from the initial state  $|2p0\rangle$  to the final states with even  $z$ -parity) up to  $w = 50$ . Without electric field ( $\tilde{F} = 0$ ) two recurrences occur at  $\tilde{S} = 0.872$  for the

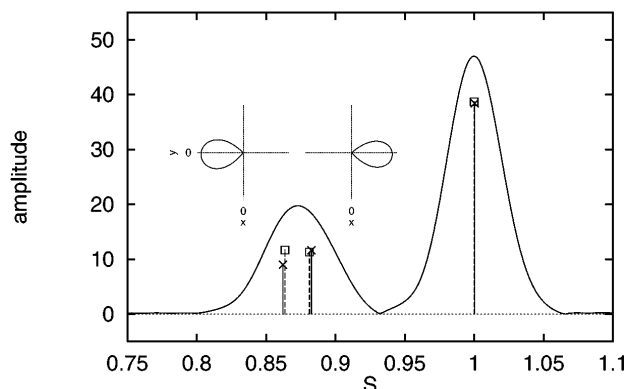


FIG. 3. Recurrence spectrum for the photoabsorption cross section of the hydrogen atom in crossed magnetic and electric fields at scaled energy  $\tilde{E} = E\gamma^{-2/3} = -0.5$  and field strength  $\tilde{F} = F\gamma^{-4/3} = 0.02$ . Transition  $|2p0\rangle \rightarrow |\pi_z = +1\rangle$ . Smooth line: Conventional Fourier transform. Solid stick spectrum and crosses: High resolution quantum recurrence spectrum. Dashed sticks and squares: Recurrence spectrum from semiclassical *closed orbit* theory. The two recurrence peaks around  $\tilde{S} = 0.87$  are identified by closed orbits in the  $(x, y)$  plane.

perpendicular orbit and at  $\tilde{S} = 1$  for the parallel orbit. With the weak electric field  $\tilde{F} = 0.02$  the parallel orbit is not significantly affected but the perpendicular orbit splits into two closed orbits with slightly different classical actions  $\tilde{S}^{\text{cl}} = 0.864$  and  $\tilde{S}^{\text{cl}} = 0.881$ . Their shapes are presented as insets of Fig. 3. As can be seen in the figure the two closed orbit contributions are not resolved by the conventional Fourier transform (smooth line) but they are clearly separated by the high resolution spectral analysis in agreement with the classical recurrence spectrum.

In summary, we have shown that interesting physical phenomena can be revealed in high resolution quantum recurrence spectra by application of the harmonic inversion technique, thereby circumventing the restrictions imposed by the uncertainty principle of the conventional Fourier transform. The method allows one, e.g., to test the validity of semiclassical theories, to identify hidden ghost orbits in the quantum spectra, and to observe the symmetry breaking in the spectra of the hydrogen atom in crossed magnetic

and electric fields. The analysis has been demonstrated here on theoretically calculated quantum spectra but may be applied to experimental spectra as well.

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