Eur. Phys. J. D **44**, 47–50 (2007)
DOI: 10.1140/epjd/e2007-00165-x

THE EUROPEAN PHYSICAL JOURNAL D

On the effect of resolution on nearest-neighbour level spacings in atomic spectra

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Received 7 March 2007
Published online 23 May 2007 –

Abstract. We discuss the effects of experimental resolution on the analysis of nearest neighbour energy level spacings for the signatures of underlying classically chaotic electron dynamics. Through a numerical treatment we arrive at a new dimensionless resolution criterion which must be met in order that statistical studies of this kind be considered meaningful.

PACS. 32.80.Rm Multiphoton ionization and excitation to highly excited states – 32.60.+i Zeeman and Stark effects

The question of the role played by chaotic trajectories in the classical limit of quantum mechanics was first raised by Albert Einstein in 1917 [1], and remains an open problem to this today. However, since quantum mechanics is now generally accepted, it would seem appropriate to rephrase Einstein's original question and rather then phrasing it as a criticism of quantum mechanics as Einstein did, to instead ask how does chaos emerge when an appropriate quantum system is taken to the classical limit? Thus the problem becomes one of how to formulate the correspondence principle rather than a test of quantum mechanics itself, and provides the motivation for much current research, both experimental and theoretical, on a range of quantum systems in their classical limit.

Several physical situations exist in which one may wish to search experimentally for quantum signatures of chaos, for example molecular systems [2], atomic systems without external fields [3], atoms in strong magnetic fields [4–6], atoms in crossed electric and magnetic fields [7–9], etc. In all of these situations, and in tests for underlying chaos in quantum computation [10], nearest neighbour level spacing statistics [11,12] are used as the experimental test. However, in each of the cases mentioned it is necessary to consider the effect that experimental resolution or computational accuracy may have on the significance of the results. This point was not listed explicitly in the set of criteria, presented previously by Connerade [13], by which one may identify suitable systems in which to study quantum chaos, and although Michaille and Pique [14] addressed the issue, a complete treatment was not given. In the present letter, we approach the problem of resolution from the perspective of the Brody test and more generally from that of the ξ-parameter [9], which we recently introduced in the analysis of the spectra of Rydberg atoms in crossed electric and magnetic fields. We show how a general minimum resolution criterion can be formulated which applies to all of the above mentioned situations.

The problem which we aim to address is related to the question: how does one recognise the emergence of chaos in experimental spectra? Having identified an experimentally accessible quantum system with underlying chaotic dynamics that fulfills the relevant criteria previously listed by Connerade [13], one must aim to study this system as close to the classical limit as possible. Under such conditions, the emergence of Wigner nearest-neighbour level spacing statistics [15] is a reasonable indicator of underlying chaotic dynamics for a three-dimensional system [16,17]. However, in order to choose an appropriate test for Wigner statistics, it is necessary to have some knowledge of the nearest-neighbour level spacing distribution function (NND) representative of the regular system from which the chaotic dynamics evolve. If the regular system is composed of states with mixed total angular momentum and parity, which begin to interact with each other as the dynamics become chaotic, the initial NND is Poisson and the Brody test [18] is appropriate. On the other hand if the regular system is composed of states with the same total angular momentum and parity (a simple sequence), as can be the case in the photoabsorption spectra of Rydberg atoms in pure magnetic fields [19] or crossed electric and magnetic fields [20,21], the associated NND is not necessarily Poisson and an alternative test must be employed.

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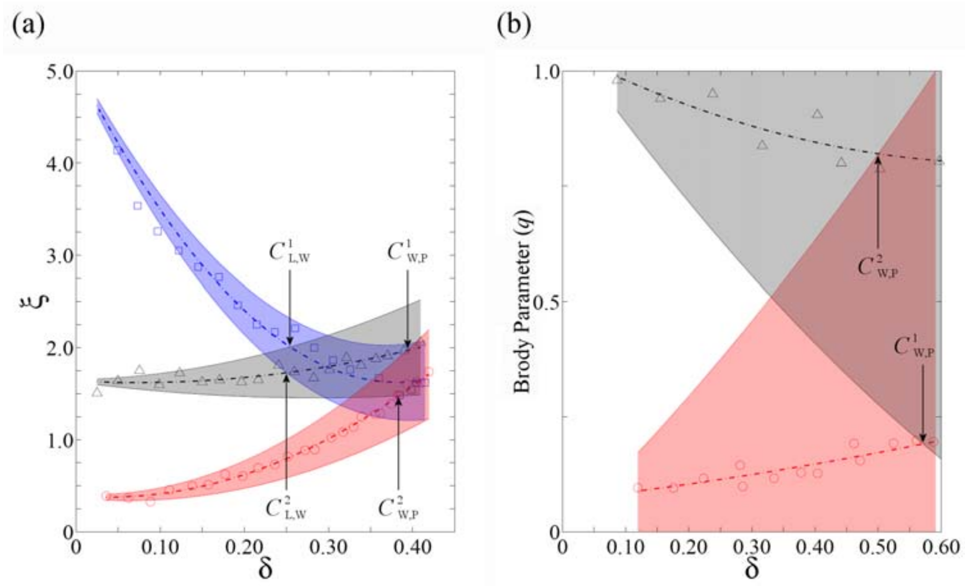

Fig. 1. (Color online) The effect of resolution on; (a) the ξ-parameter for Poisson (blue-□), Wigner (black-△) and Lorentzian (red-○) distribution functions, and (b) the Brody parameter, q, for Poisson (blue-□) and Wigner (black-△) distribution functions. The heavy dashed lines in the centre of the shaded regions in each figure are a least squares second-order polynomial fit to the calculated values of ξ and q at each resolution, with the error indicated by the range of the shaded regions surrounding each curve. The points labelled $C^i_{\mathrm{L,W}}$ and $C^i_{\mathrm{W,P}}$, where $i = 1, 2$, at which the error limits on one curve cross the central fitted curve for the adjacent distribution are those from which the resolution criterion have been determined.

To deal with the problem of identifying NNDs outside the range in which the Brody test is applicable, particularly under conditions in which the spectra with underlying regular dynamics exhibit a 'picket-fence' character (i.e. equally spaced levels), we recently introduced a new dimensionless parameter of the NND, $\xi = m/\sigma$, where m is the mode of the distribution and σ is the standard deviation from the mean [9]. The use of this ξ-parameter permits the identification of NNDs arising from 'picket-fence' type spectra (typically Lorentzian or Gaussian depending upon the instrument function associated with a particular experiment), and those which are Wigner or Poisson.

In order to provide a basis with which to establish a criterion for distinguishing between Poisson, Wigner and Lorentzian distributions, we have generated model data sets corresponding to each, under a range of resolution conditions, and analysed them in terms of the ξ-parameter. These data sets, each composed of 200 spacings (typical of the spectra reported in Ref. [9]), were generated randomly using a rejection method [22]. Unresolved spacings under each set of resolution conditions were removed, divided by two, and each half was added to two randomly selected points remaining in the data set, thus reproducing the effect of finite experimental resolution. The minimum resolution was initially set in units of 'true' mean level spacing, in the range from 0.025 to 0.5, and following the generation of each model data set, was recalculated in units of the new 'measured' mean level spacing resulting in the ratio (δ) of the resolution to the mean level spacing which corresponds to the FWHM of the instrument function associated with the experiment in units of 'measured' mean level spacing.

The ξ-parameter was calculated for each of the model data sets by a method similar to that described in reference [9]. For each data set, expressed in units of mean level spacing, the standard deviation from the mean could be calculated directly, with its associated error a function of the size of the dataset. The mode was then calculated by first determining the minimum appropriate histogram bin-width, and then shifting the positions of the bin-centres in steps of 1/100th of the bin-width across a full bin-width while finding the value of the mode at each step. The average mode for each data set, and its associated error (a function of both the bin-width and the standard deviation from the mean) could then be determined.

The crucial step in this process, as with many other methods of determining the form of a statistical distribution, including the Brody test, is the choice of bin-width. As we are dealing with measurements of level spacings made at finite resolution, *the minimum meaningful bin-width which can be employed is equal to* δ, the FWHM of the instrument function in units of mean 'measured' level spacing.

From the analysis of the average behaviour of 20 model data sets for each distribution at each resolution, we have been able to construct a picture, presented in Figure 1a, of the effect of finite resolution on the possibility to distinguish between Poisson, Wigner and Lorentzian NND's using the ξ-parameter. From this figure, it is clear that δ, has a very significant impact on the measured value of ξ. If the FWHM of the instrument function is not sufficiently fine (right hand side of Fig. 1a) all of the distributions overlap and cannot be separated from each other. If on the other hand δ is small they can be readily distinguished.

The shaded areas above and below each of the Poisson, Wigner and Lorentzian curves indicate the error on ξ at each resolution. Thus the point at which the error limits on one curve cross the central fitted curve for the adjacent distribution (labelled $C^i_{\mathrm{L,W}}$ and $C^i_{\mathrm{W,P}}$, where $i = 1, 2$) is where the two distributions can no longer be distinguished. It is these points that then provide us with the resolution criteria that we seek. Thus from Figure 1a, the maximum value of δ for which one can distinguish between Lorentzian and Wigner distributions (i.e. the mean of the points labelled $C^1_{\mathrm{L,W}}$ and $C^2_{\mathrm{L,W}}$) is $\sim$0.25, and between Poisson and Wigner distributions (i.e. the mean of the points labelled $C^1_{\mathrm{W,P}}$ and $C^2_{\mathrm{W,P}}$) is $\sim$0.30.

It is important to note that δ, the FWHM of the instrument function which is plotted, is a dimensionless parameter as it is referred to the mean level spacing of the spectrum, thus the appropriate resolution to be used in an experiment of this kind depends very much on the level density. For example, as one approaches the classical limit, in an experiment using Rydberg atoms, it may well be that the available resolution becomes insufficient to distinguish between the different types of distribution. The curves in Figure 1a therefore provide a simple and general criterion to determine, before conducting such an experiment, whether the spectral resolution available will be adequate.

The above analysis of the effect of resolution on the ξ-parameter when testing for the presence of chaotic underlying dynamics in a quantum system can also be extended to the Brody test. To do this we have generated model Poisson and Wigner distributions as above, each composed of 200 spacings, and again carried out the necessary matching of the minimum bin-width to the spectral resolution. The results of this calculation are presented in Figure 1b. In this case, the error on the Brody parameter, q, was obtained by determining q for each binned histogram, for the set of bin-centres along with the sets of low-spacing and high-spacing edges of each bin, giving negative and positive errors respectively on the measured value of q. Thus under conditions in which a regular spectrum is expected to display Poisson nearest neighbour spacing statistics which follow the evolution of the Brody function toward Wigner statistics as the dynamics become chaotic, the limiting value of δ (i.e. the mean of the points labelled $C^1_{\mathrm{W,P}}$ and $C^2_{\mathrm{W,P}}$) is $\sim$0.5. A similar result to this was obtained previously by Michaille and Pique [14], however by overlooking the limit imposed by the spectral resolution on the minimum allowed histogram bin-widths in their work, accurate determination of the error on q and hence the reliability of q itself could not be achieved.

In the examples of chaotic spectra given by Connerade [13], the levels concerned all belong to autoionising spectra, in which case the instrumental resolution is much finer than the autoionisation linewidth and it follows that instrumental resolution does not limit the ability to separate the distributions from each other. Rather it is the autoionisation width, intrinsic to these spectra, which acts as the fundamental limitation. This width corresponds to the linewidth in Figure 3b of Connerade [13] and is seen to be considerably smaller than the average spacing in that spectrum. Consequently the examples given by Connerade [13] lie towards the left hand side of Figure 1 in the present paper, where it is possible to distinguish experimentally between Lorentzian, Wigner and Poisson distributions. It is also important to note that in the first autoionising range the widths of the lines scale as $1/n^3$ which means that the ratio of the linewidth to the level spacing for field-free Rydberg spectra is a constant. Thus one can extend measurements to higher and higher n-values without migrating to the right hand side of Figure 1 provided the instrumental resolution is always much better than the autoionising linewidth. However, this will not be true as one enters the energy range where Auger broadening occurs. In the latter case, the broadening does not scale as $1/n^3$, but remains constant and moving up in n-value inevitably leads to moving across Figure 1 towards the right. This situation was considered by Connerade [13] who described it as a truncation of the Rydberg series which would not allow the classical limit to be reached. The present criterion is more quantitative and sets a more specific limit beyond which Auger broadening would prevent the observation of a Wigner distribution.

The problem discussed in the present letter arises primarily when one tries to extend experimental observations to high Rydberg members in the spectral region below the first ionisation potential. In this case it is instrumental broadening which sets the limit for recognising the emergence of a Wigner distribution and any spectrum of this type must be observed under conditions which satisfy the criterion of Figure 1. For instance we have recently reported experiments on atoms in crossed-fields at n-values between 52 and 55. These spectra were observed under conditions which correspond to a ratio of resolution to mean level spacing of $\delta \simeq 0.26$. Under these conditions, at the limit of the resolution criterion established above for the ξ-parameter, the possibility to distinguish a Wigner distribution from a Lorentzian distribution is severely limited and at best a tendency for the distribution to change from Lorentzian to Wigner may be identified. It is therefore clear that it would not be possible to extend the observations to much higher values of n using the same excitation and detection scheme without intruding into the right hand side of Figure 1a where the three NNDs can no longer be separated from each other. Thus any further experiments will require a substantial increase in spectral resolution above that provided by our combination of magnet, single mode, injection seeded OPO laser and excited state detection scheme.

It is hoped that the diagrams in Figure 1 and the criteria reported here will be of use in the design and analysis of future experiments on nearest level spacings and their statistics.

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