Experimental Study of Energy-Level Statistics in a Regime of Regular Classical Motion

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We have studied energy-level statistics of the diamagnetic lithium Rydberg spectrum in a regime of regular classical motion. The distribution of adjacent levels displays the Poisson-type behavior that has been reported in theoretical studies of numerous systems. Long-range correlations in the spectrum can be related simply to the number of underlying manifolds.

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Energy-level statistics are believed to be an important signature of classical dynamics in quantum systems.[1] In particular, it is generally accepted that regular classical dynamics for a nonseparable Hamiltonian is manifested by Poisson statistics of energy-level positions. In contrast, systems that display irregular or chaotic classical motion are characterized by the statistics of random-matrix eigenvalue ensembles. The classical dynamics of the diamagnetic hydrogen atom is known to display both regular and irregular behavior,[2,3] and quantum calculations reveal that the spectral statistics display the corresponding expected behavior.[3,4] Thus, the diamagnetic structure of hydrogen and similar Rydberg atoms provides an ideal testing ground for investigation of the manifestations of nonlinear classical behavior in a quantum system. We report here what to our knowledge is the first experimental study of energy-level statistics in a regime of classically regular motion. The distribution of spacings between adjacent energy levels is in general agreement with predictions. We have also studied correlations in the spectrum. For short range, these correlations are Poisson type. At long range, however, there is a clear departure from Poisson behavior that can be related simply to the number of underlying manifolds.

The Hamiltonian for the hydrogen atom in a magnetic field $B\hat{\mathbf{z}}$ can be written

$$H=\frac{p^2}{2}-\frac{1}{r}+\frac{1}{2}L_zB+\frac{1}{8}B^2(x^2+y^2)\,, \tag{1}$$

where the unit of magnetic field is

$$m^2e^3(4\pi\epsilon_0)^{-2}\hbar^{-3}=hcR_\infty/\mu_B=2.35\times10^5\ \mathrm{T}\,.$$

Relativistic effects and interactions of the electron and nuclear spins are neglected. The classical mechanics of this system is conveniently expressed in terms of scaled variables: $\tilde{r}=B^{2/3}r$ and $\tilde{p}=B^{-1/3}p$. It is readily shown that $H=B^{2/3}\tilde{H}$, where

$$\tilde{H}=\frac{\tilde{p}^2}{2}-\frac{1}{\tilde{r}}+\frac{1}{2}\tilde{L}_z+\frac{1}{8}(\tilde{x}^2+\tilde{y}^2)\,. \tag{2}$$

Because $\tilde{H}$ does not depend on B, the full range of classical behavior is characterized by the parameters $\tilde{L}_z$ and $\tilde{E}$. Numerical studies[3] of the trajectories governed by $\tilde{H}$ reveal orderly motion for $\tilde{E}<-0.54$. In the regime $-0.54\le\tilde{E}\le-0.127$, both regular and irregular trajectories are found, and for $\tilde{E}>-0.127$, the motion appears to be fully chaotic. These regions are displayed in Fig. 1.

Studying, energy-level structure experimentally requires surveying an energy range containing a sufficient number of eigenstates to provide good statistics. The range available for studying regular statistics is limited. At zero field the energy levels form a Rydberg series which characterizes purely periodic motion. At low fields each term splits into a manifold of levels that initially evolve as parabolas. As the field increases these manifolds overlap, as shown in Fig. 2. Poisson-type behavior requires the superposition of many manifolds. Figure 2 shows that to have many manifolds mixed and also to be in the classically regular regime, it is desirable to work with high E and low B. However, the energy-level density also increases with E. Consequently, finite spectral resolution places an upper limit on the energy

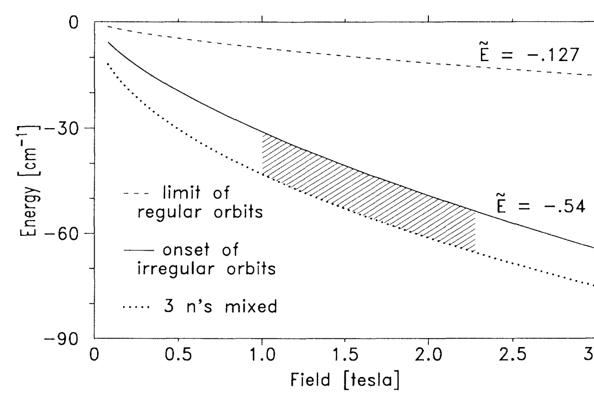

FIG. 1. Regions of differing behavior of a Rydberg atom in a magnetic field. Classical orbits below the solid line are regular; above the dashed line the orbits are believed to be chaotic. The dotted line indicates where the highest-lying level of manifold n crosses the lowest-lying level from manifold $n+3$. The shaded area is the region studied in this work.

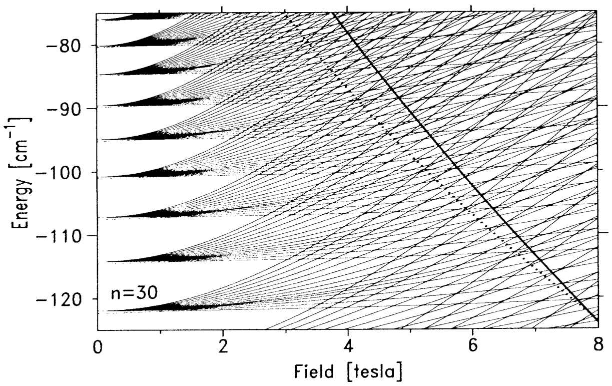

FIG. 2. Calculated eigenvalue map for lithium $m=0$ odd-parity states. The solid and dotted lines are the same as in Fig. 1.

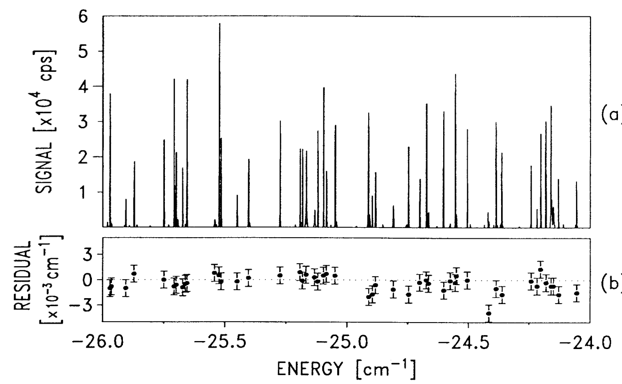

FIG. 3. (a) Sample of the data. This 2-cm^{-1} section of the spectrum, taken at 1.0797(2) T, showing 48 fully resolved spectral lines in the regular regime, is typical. A total of 871 levels were measured over a range of ~ 20 cm^{-1} at each of three different fields. (b) The difference between the calculated and measured position for each line in (a). The error bar is due to the limited step of the laser sweep. The energy is relative to the zero-magnetic-field ionization limit.

range that can be explored reliably. The shaded region in Fig. 1 shows the range that we have selected for study.

Experimental studies of Rydberg atoms in high magnetic fields have been carried out by several groups[5-7]; however, none has reported results with the resolution needed to study energy-level statistics in the regular regime. Lithium, in contrast, can be studied with high-resolution cw laser methods. Furthermore, the states which we have selected—odd parity—are hydrogenlike in their statistics: The largest quantum defect, $\delta_p=0.05$, has only a minor effect on the spectrum, as will be discussed below. (Even-parity $m=0$ states are less hydrogenlike because of the s-state quantum defect, $\delta_s=0.40$.)

The general experimental approach is similar to that of an earlier study by our group,[8] with the important advance of the introduction of cw lasers and precision optical metrology. A highly collimated atomic beam of lithium traveling parallel to the magnetic field is excited to Rydberg states by a two-step process: the $2S\rightarrow 3S$ two-photon transition, and $3S\rightarrow$ Rydberg state. The resolution is 10^{-3} cm^{-1} FWHM. The absolute accuracy of all lines observed is $\pm 2\times 10^{-3}$ cm^{-1}; the relative precision is set by the resolution. The magnetic field is determined by exciting $|\Delta m|=1$ transitions and observing the paramagnetic splitting of the $n=21$ manifold.[9] Magnetic fields in the range of 1-2 T were used; the uncertainty in field is $\pm 2\times 10^{-4}$ T. Details of the experiment will be published elsewhere.

We have also calculated the spectrum for the regime reported here. This was done to provide a consistency check on our experiments, giving confidence in our methods for applications in regimes where calculations cannot be performed, and to verify our calculational method. Matrix elements of the Hamiltonian are computed in a spherical basis by techniques described previously.[10] The matrix is diagonalized at each field point. A basis of $n=2$-100, $m=0$, odd-parity states is used. (The total number of states is 2500.) The results agree with experiment to within the experimental accuracy. A sample of the data and comparison with calculations are shown in Fig. 3.

Spectra were taken at 1.1, 1.2, and 2.3 T. With use of a second-order polynomial fit, the energy scales were linearized for each spectrum to remove small global variations in the density of states, and normalized to unit mean spacing. Histograms of the separation between adjacent levels were constructed, and added. The result is shown in Fig. 4. This procedure assumes a lack of correlations between the separate spectra which we believe to be justified by the large number of level crossings between them. Results for the calculated spectrum are also shown in Fig. 4. The total number of experimental levels is 391, while the total number of calculated levels is 396. The difference is due to missed levels at anticrossings in-

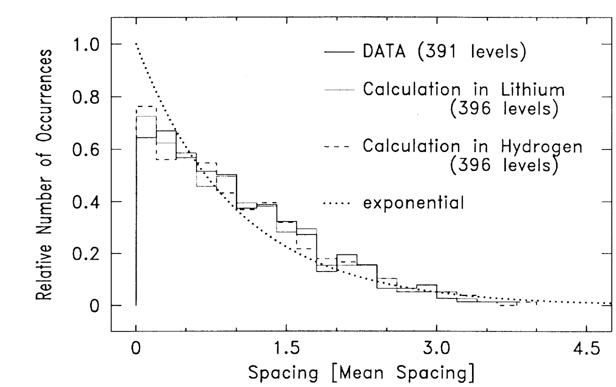

FIG. 4. Adjacent-energy-level spacing histogram. The approximately exponential behavior characteristic of Poisson statistics is evident. The depression of the first bin of data is due to the missed levels, all of which were at small spacings.

duced by the lithium core. At these anticrossings all the oscillator strength is transferred to one state, rendering the other observable with our excitation scheme.[11] The result is fewer levels at small spacings.

To investigate the difference between the lithium and hydrogen spectra, we have also calculated the eigenvalues for hydrogen in the regime under study. Small differences in the spectrum are apparent due to anticrossings induced by the lithium core, but their effect on the level-spacing histogram is minor. The average anticrossing size is much less than the mean spacing, and the anticrossings affect only the first bin of the histogram (see Fig. 4). Much more significant than the difference between lithium and hydrogen, however, is the difference between either histogram and an exponential curve. As pointed out by Delande and Gay,[3] this discrepancy is a manifestation of the fundamentally nonrandom nature of a spectrum which has underlying periodicities.

Long-range correlation between levels provides a complementary picture of energy-level distributions. A widely used measure is the $\bar{\Delta}_3$ statistic of Dyson and Mehta.[12] $\bar{\Delta}_3(L)$ is the average mean squared deviation per unit energy interval between the energy-level accumulation function (the "staircase" function) and the best-fit straight line, where the average is over intervals of L mean spacings. For Poisson specta, totally uncorrelated, $\bar{\Delta}_3(L)=L/15$. For correlated spectra, $\bar{\Delta}_3(L)$ increases less rapidly with L. For Gaussian-orthogonal-ensemble statistics,[13] $\bar{\Delta}_3(L)$ increases asymptotically as $\ln(L)$; for a "picket fence" spectrum, $\bar{\Delta}_3(L)$ is constant for large L and equal to $\frac{1}{12}$.

Figure 5 shows $\bar{\Delta}_3(L)$ for our data. It can be seen that $\bar{\Delta}_3(L)$ initially increases as $L/15$, but departs from this Poisson-type behavior and appears to approach a value of roughly 0.4, indicating long-range correlations in the spectrum. For some systems this type of behavior can be related to the "break time"—that is the period of the shortest closed classical orbit.[14] We have found that the observed behavior can be related simply to the number of terms whose diamagnetic manifolds contribute to the spectrum. This approach avoids the consideration of classical orbits.

To study the effect of the superposition of manifolds, we have calculated $\bar{\Delta}_3$ for a series of superimposed incommensurate picket-fence spectra. As shown in Fig. 6(a), $\bar{\Delta}_3$ approaches the limiting value of $N/12$, where N is the number of series. This merely illustrates the additive nature of $\bar{\Delta}_3$ for superpositions of independent spectra.[15] To our surprise, however, we found the same behavior for a series of diamagnetic manifolds generated for hydrogen, ignoring n mixing. As can be seen in Fig. 2, these manifolds are by no means equally spaced. Nevertheless, the single limiting behavior $\bar{\Delta}_3 \sim N/12$, where N is the number of manifolds, appears to hold accurately.

Our data were taken over a regime in which varying numbers of manifolds contribute. The limiting value, $\bar{\Delta}_3 \sim \frac{5}{12}$, indicates that the mean number of underlying manifolds is ~ 5. This value is consistent with observations based on our calculated eigenvalue map. To obtain a larger number of levels and contributing manifolds, we have also extended the scans somewhat into the slightly irregular regime. Figure 5 also shows the $\bar{\Delta}_3$ statistic for these data and the corresponding calculation. The limiting value of $\bar{\Delta}_3 \sim \frac{7}{12}$, implying ~ 7 contributing manifolds, is also consistent.

We believe that this experiment provides a credible illustration for the Poisson-type statistics of spectra in the

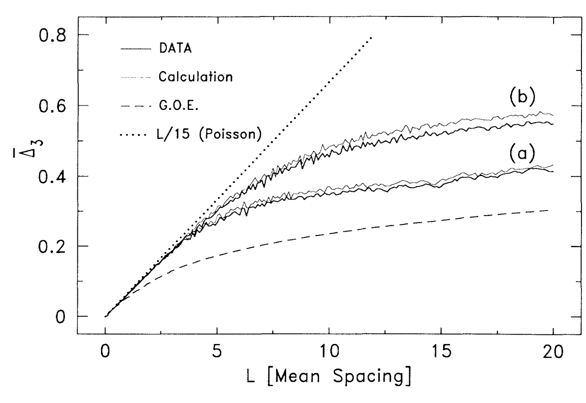

FIG. 5. The $\bar{\Delta}_3$ statistics for our data (solid lines) showing Poisson-type behavior for small L. Curves a, data strictly in the regular regime shown in Fig. 1 (391 levels measured; 396 levels calculated). Curves b, data in and slightly above the regular regime (871 levels measured; 890 levels calculated). The saturation of $\bar{\Delta}_3$ with limiting values of approximately 0.4 and 0.6, respectively, is a manifestation of a small number of contributing manifolds.

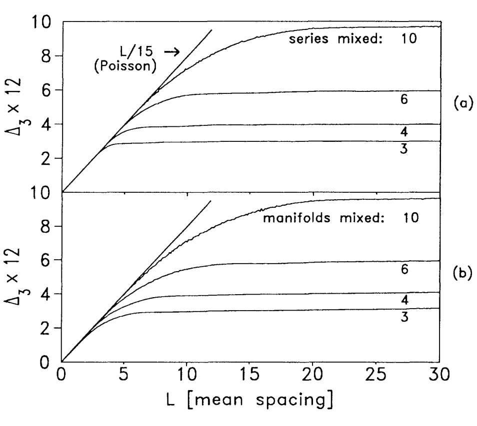

FIG. 6. Demonstration of the additive nature of $\bar{\Delta}_3$. (a) Superposition of series of equally spaced lines. The limiting value of $\bar{\Delta}_3$ is $N/12$, where N is the number of series mixed. (b) $\bar{\Delta}_3$ of calculated spectra for hydrogen, ignoring n mixing, in regions where a well determined number of manifolds have crossed. The limiting value of $\bar{\Delta}_3$ is again $N/12$, where N is the number of manifolds which have crossed.

regime of orderly classical motion. Having approached the problem experimentally, however, the question naturally arises as to what conclusions about the nature of the Hamiltonian can be drawn from a given data set. The adjacent-neighbor spacing distribution provides some indication of the degree of level repulsion, but in practice obtaining a statistically significant number of levels presents a considerable experimental challenge. Although it is accepted that long-range correlations indicate chaotic classical behavior, these can also arise in spectra comprised of a limited number of orderly manifolds. Experimentally distinguishing between the various types of limiting behavior is thus likely to be difficult. Considering the large body of accurate data that is required for meaningful tests of spectral statistics and the relatively modest conclusions that can be drawn, one is naturally led to ask whether more efficient methods may exist for using the spectral information.

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[1]*Statistical Theories of Spectra: Fluctuations,* edited by C. E. Porter (Academic, New York, 1965); M. V. Berry and M. Tabor, Proc. Roy. Soc. London A **356**, 375 (1977); O. Bohigas and M. J. Giannoni, in *Mathematical and Computational Methods in Nuclear Physics,* edited by J. S. DeHesa, J. M. G. Gomez, and A. Polls, Lecture Notes in Physics Vol. 209 (Springer-Verlag, New York, 1984).

[2]M. Robnik, J. Phys. A **14**, 3195 (1981); A. Harada and H. Hasegawa, *ibid.* **16**, L259 (1983); J. B. Delos, S. K. Knudson, and D. W. Noid, Phys. Rev. A **30**, 1208 (1984).

[3]D. Delande and J. C. Gay, Phys. Rev. Lett. **57**, 2006 (1986); G. Wunner, U. Woelk, I. Zech, G. Zeller, T. Ertl, F. Geyer, W. Schweitzer, and H. Rudder, *ibid.* **57**, 3261 (1986).

[4]D. Wintgen and H. Friedrich, Phys. Rev. Lett. **57**, 571 (1986).

[5]P. Cacciani, E. Luc-Koenig, J. Pinard, C. Thomas, and S. Liberman, Phys. Rev. Lett. **56**, 1124 (1986); J. Phys. B **19**, L519 (1986).

[6]A. Holle, G. Wiebusch, J. Main, K. H. Welge, G. Zeller, G. Wunner, T. Ertl, and H. Ruder, Z. Phys. D **5**, 279 (1987); A. Holle, J. Main, G. Wiebusch, H. Rottke, and K. H. Welge, Phys. Rev. Lett. **61**, 161 (1988).

[7]J. Neukammer, H. Rinneberg, K. Vietzke, A. König, H. Hieronymus, M. Kohl, and H.-J. Grabka, Phys. Rev. Lett. **59**, 2947 (1988).

[8]M. L. Zimmerman, J. C. Castro-Neto, and D. Kleppner, Phys. Rev. Lett. **40**, 1083 (1978); J. C. Castro, M. L. Zimmerman, R. G. Hulet, D. Kleppner, and R. R. Freeman, *ibid.* **45**, 1780 (1980).

[9]M. M. Kash, G. R. Welch, C. Iu, and D. Kleppner, Spectrochemica Acta A (to be published).

[10]M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A **20**, 2251 (1979).

[11]Cacciani *et al.*, Ref. 5 (J. Phys. B).

[12]F. J. Dyson and M. L. Mehta, J. Math. Phys. **4**, 701 (1963).

[13]M. L. Mehta, *Random Matrices and the Statistical Theory of Energy Levels* (Academic, New York, 1967).

[14]M. V. Berry, Proc. Roy. Soc. London A **400**, 229 (1985).

[15]A. Pandey, Ann. Phys. (N.Y.) **119**, 170 (1979).