Orderly Structure in the Positive-Energy Spectrum of a Diamagnetic Rydberg Atom

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We have observed orderly structure in the spectrum of lithium in the range $-1 \leq E \leq \pm 8$ cm^{-1}, in magnetic fields of approximately 6 T. The structure resembles series of Rydberg levels converging on Landau levels. Interactions between levels are evident, but the underlying orderly structure raises hopes that a quantum solution to the problem may be feasible. The structure occurs in a regime where the classical motion is believed to be chaotic.

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We report evidence for orderly energy-level progressions in the positive-energy spectrum of a Rydberg atom in a magnetic field. Despite the simplicity of the Hamiltonian, quantum solutions in this regime have not yet been obtained. Our observations suggest that the problem may be more tractable than previously believed and that a quantum solution may be feasible. Such an achievement would represent a significant advance in atomic theory because this system is a paradigm for elementary quantum systems with nonseparable Hamiltonians.[1] The Rydberg atom in a magnetic field is also a paradigm for quantum systems whose classical motion is chaotic.[2] The orderly progressions that we observe lie in a regime where the classical motion appears to be fully chaotic.[3,4] This observation is contrary to the accepted understanding of spectral properties in a regime of disorderly motion.

A considerable literature exists on the theory of Rydberg atoms in magnetic fields, encompassing approximate analytical solutions based on group-theoretic methods, numerical computations, and semiclassical analysis. The problem is the subject of a number of reviews.[2,5–8]

The Hamiltonian for a hydrogen atom in a magnetic field along the z axis is (using atomic units and cylindrical coordinates)

$$H = \frac{p^2}{2} - \frac{1}{(\rho^2+z^2)^{1/2}} + \frac{m}{2}B + \tfrac{1}{8}B^2\rho^2\,, \qquad (1)$$

where m is the azimuthal quantum number and the unit of magnetic field is $hcR_\infty/\mu_B = 2.35\times10^5$ T. Because our experiment employs lithium rather than hydrogen, the Coulomb potential must be modified within the lithium ionic core, i.e., at distances less than a few atomic units. However, as discussed previously,[9] the largest quantum defect for an odd-parity state of lithium, δ_p, is only 0.04. This is so small that lithium's behavior in the low-energy regime of regular motion has been found to be essentially hydrogenic. Based on this observation, we surmise that the effect of the lithium core is unimportant in the positive-energy regime.

At high magnetic fields [$B \gg 1$ atomic unit (a.u.)] the problem can be viewed as a combination of rapid motion around the z axis due to the magnetic field superimposed on slow motion along the z axis due to the Coulomb potential. The result is a series of free electron states —Landau levels—each of which supports a Rydberg progression whose ground state lies above the next lower Landau level. Friedrich[10] has shown that in this regime the energy of $m=0$ states is given by

$$E(n_\rho,n_z^*) = (n_\rho + \tfrac{1}{2})B - \frac{1}{2(n_z^*)^2}\,, \qquad (2)$$

where n_ρ is a non-negative integer. The effective quantum number n_z^* can be written in terms of a quantum defect as $n_z^* = n_z - \mu$, where n_z is a positive integer. The quantum defect μ is negative because the transverse motion tends to average the Coulomb potential over ρ, reducing its effect along the field. Also, μ depends on n_ρ and on the field, but varies only slowly with the energy.

Our experiment employs an atomic beam of lithium moving parallel to the magnetic field. The Rydberg states are excited by two-photon excitation of the $2S \rightarrow 3S$ transition and single-photon excitation of odd-parity Rydberg states. The excited atoms are detected by electric field ionization. The instrumental resolution is 1×10^{-3} cm^{-1} FWHM, the uncertainty in energy is $\pm2\times10^{-3}$ cm^{-1}, and the uncertainty in magnetic field is $\pm5\times10^{-4}$ T. Figure 1 shows our results for odd-parity $m=0$ states near the ionization threshold at 6 T. The most striking features of the data are a number of widely spaced strong lines with large slopes with magnetic field and, less conspicuously, families of adjacent levels with relatively small slopes. Anticrossings and other phenomena associated with level interactions are apparent, but the overall impression is of an orderly spectrum composed of identifiable levels.

At 6 T (2.6×10^{-5} a.u.) the longitudinal and transverse motions are strongly coupled and there is no reason to believe that Eq. (2) could be usefully applied. Nevertheless, it provides a starting point based on the following considerations. The rms radius of a Landau state is $\langle\rho^2(n_\rho)\rangle^{1/2} = 2[(n_\rho+\frac{1}{2})/B]^{1/2}$. For example, at 6 T $\langle\rho^2(0)\rangle^{1/2} = 280a_0$ and $\langle\rho^2(10)\rangle^{1/2} = 1400a_0$. For the

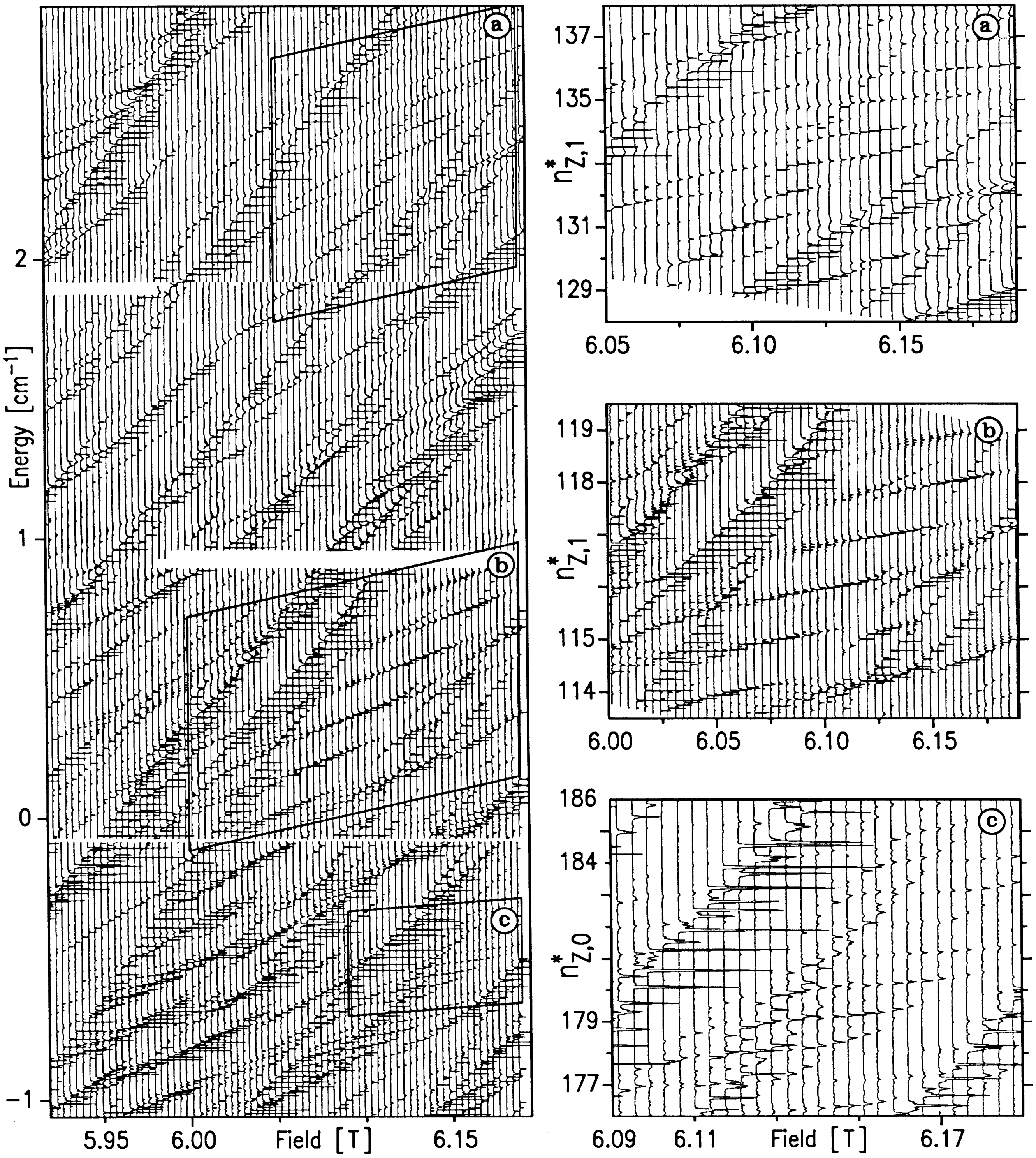

FIG. 1. Energy-level map of odd-parity $m=0$ states of lithium in an applied magnetic field. The maps are created by scanning a laser at successively higher fields: Horizontal peaks are field ionization signals. (The gaps are regions which were skipped during data collection.) The structure is most easily seen by viewing close to the plane from the left. Reduced-term-value plots for the outlined regions are shown at the right. (a), (b) $n_\rho=1$; (c) $n_\rho=0$. As explained in the text, with these coordinates a progression of Rydberg states appears as a series of lines with unit separation. The ionization threshold occurs at approximately $+2.8$ cm^{-1}.

one-dimensional Coulomb problem,[11] $\langle z^2\rangle^{1/2} \approx n_z^2$. Thus, for $n_\rho \leq 10$ and $n_z \gtrsim 100$, we find $\langle z^2\rangle^{1/2} \gg \langle\rho^2\rangle^{1/2}$. In this limit $\langle\rho^2+z^2\rangle^{1/2} \approx \langle z^2\rangle^{1/2}$ and Eq. (1) can be decomposed into the Hamiltonian for a free electron in a magnetic field plus that of a one-dimensional hydrogen atom. Under such conditions the energy levels consist of interleaved families of Rydberg progressions, one progression converging to each Landau level. However, interactions between these levels due to the mixing of transverse and longitudinal motion near the nucleus could destroy all semblance of order. Thus the orderly features of our data are unexpected. We emphasize that this argument is intended to provide a framework for interpreting the important features of the data: It is not intended for a quantitative understanding of the system.

Taking Eq. (2) as a starting point, let us turn to the data. The slope of energy with magnetic field of an $m=0$ Landau level[9] is $\partial E/\partial B = n_\rho + \frac{1}{2}$. The strongest line in Fig. 1 has a slope of 8.8 cm^{-1}/T, which corresponds to $n_\rho \approx 9$. If n_ρ were 9, then the energy of the level implies that $n_z \approx 46$. The relatively high oscillator strength of the high-slope states implies that they are concentrated in the vicinity of the nucleus, in contrast to states with low value of n_ρ and much higher values of n_z. To enhance the features of the high Rydberg states we have removed the global effect of the magnetic field by plotting the data in terms of an effective quantum number, n_z^*. Using the notation $n_{z,j}^*$, where $j=n_\rho$, we have

$$n_{z,j}^* \equiv [(2j+1)B-2E]^{-1/2}. \qquad (3)$$

In a reduced term value plot, a plot of $n_{z,j}^*$ vs B, a progression of Rydberg levels appears as a series of levels *with unit separation.* If the quantum defect is constant, the levels have zero slope.

The data in Fig. 1 show numerous fragments of Rydberg progressions. Three regions have been extracted and displayed as reduced-term-value plots. Insets (a) and (b) show portions of the progression converging on $n_\rho=1$; inset (c) shows part of a progression converging on $n_\rho=0$. Evidence that these are indeed Rydberg progressions is provided by the unit spacing between the levels. However, it is also evident that the slopes are not zero: The quantum defect varies significantly with B across the plot. This is a clear indication that application of Eq. (2) at these fields is unrealistic. Furthermore, the positive slope of n_z^* indicates that the magnitude of the (negative) quantum defect μ increases with B, whereas one would expect the magnitude to decrease as the system becomes more one dimensional. In addition, interactions with higher n_ρ states are significant. Anticrossings are evident, though they are often sharp [as in inset (b); $B=6.05$ T, $n_{z,1}^*=115.5$], and the oscillator strengths show broad variations.

Figure 1 displays part of a larger set of data that we have taken. Most of the data in Fig. 1 lie below the ionization (i.e., lowest Landau energy), which at a field of 6 T occurs at +2.8 cm^{-1}. However, as we have recently shown,[9] structure continues through the ionization limit (though presumably adequate sensitivity would reveal a converging Rydberg progression). Figure 2 shows a portion of the data near +7 cm^{-1}, illustrating a Rydberg progression for $n_\rho=2$.

The signatures of level interactions in our data are to be expected: What is unexpected is the very existence of orderly progressions. They indicate that although the problem is nonseparable, the mixing between transverse

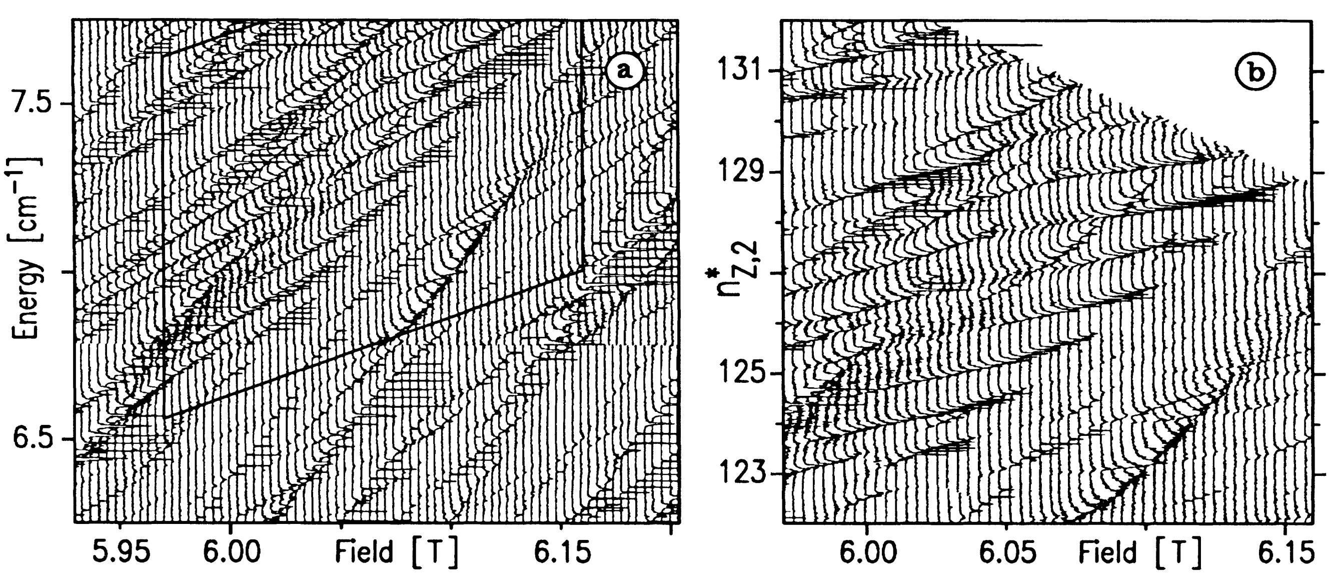

FIG. 2. (a) Same as Fig. 1 for energy near +7 cm^{-1}. (b) Reduced-term-value plot: $n_\rho=2$.

and longitudinal motion is not strong enough to destroy all order, at least in the region studied. This encourages the hope that comprehensive theoretical solutions to this problem may be feasible. It should be borne in mind, however, that the progressions we have observed all involve high values of n_z. As n_z decreases, level interactions can be expected to increase and eventually n_z and n_ρ should lose their usefulness as approximate quantum numbers.

Let us now address the data from the point of view of nonlinear dynamics. The classical motion of the diamagnetic Rydberg atom is known to evolve from regular to irregular as the energy is increased in a fixed field.[12] The motion appears to become completely irregular at an energy[3] $E_c = -0.127B^{2/3}$. At 6 T, $E_c = -24\ \text{cm}^{-1}$. Calculations of the energy-level structure reveal that the spectral fluctuations evolve in the expected fashion as one progresses from the regime of orderly motion to the regime of disorderly motion.[3,4] (Experimental evidence for the spectral fluctuations in the orderly regime was the subject of an earlier work.[13]) Although we have not attempted a statistical analysis of our data, it is apparent that the underlying order is contrary to the random-matrix behavior of eigenvalues that has been observed in many model systems.[14]

In interpreting these data, the possible effect of the stray electric field must be considered. Typically, the electric field has a magnitude of $0.025\ \text{V}\,\text{cm}^{-1}$ and is directed primarily along the z axis. This is about half the value of the stray field of the data presented in Ref. 9. There is a slight narrowing of some of the spectral lines compared to our earlier data, but the overall agreement between the two sets of data is excellent. Thus, although we believe that the electric field causes observable effects, it does not appear to play a dominant role.

A possible explanation is that stable orbits exist in the regime of our studies, but have so far eluded calculation. Such a finding would suggest, at the least, that the transition from regular to irregular motion is subtler than recognized. As a last resort, we would have to consider revising our understanding about the manifestations of disorderly motion in quantum systems.

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[1]U. Fano, Comments At. Mol. Phys. **22**, 97 (1988).

[2]H. Hasegawa, M. Robnik, and G. Wunner, Suppl. Prog. Theor. Phys. (to be published).

[3]D. Delande and J. C. Gay, Comments At. Mol. Phys. **19**, 35 (1986).

[4]G. Wunner, U. Woelk, I. Zech, G. Zeller, T. Ertl, F. Geyer, W. Schweitzer, and H. Rudder, Phys. Rev. Lett. **57**, 3261 (1986); D. Wintgen and H. Friedrich, Phys. Rev. Lett. **57**, 571 (1986).

[5]J. C. Gay, in *Atoms in Unusual Situations,* edited by J. P. Briand (Plenum, New York, 1986), p. 107; in *Spectrum of Atomic Hydrogen,* edited by G. W. Series (World Scientific, Singapore, 1988).

[6]G. Wunner and H. Ruder, Phys. Scr. **T36**, 291 (1987).

[7]H. Friedrich and D. Wintgen, Phys. Rep. (to be published).

[8]A. R. P. Rau, Rep. Prog. Phys. (to be published).

[9]G. R. Welch, M. M. Kash, C. Iu, L. Hsu, and D. Kleppner, Phys. Rev. Lett. **62**, 1975 (1989).

[10]H. Friedrich, Phys. Rev. A **26**, 1827 (1982).

[11]R. Loudon, Am. J. Phys. **27**, 649 (1959). Note that odd-parity states have $2n_z - 1$ nodes.

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

[13]G. R. Welch, M. M. Kash, C. Iu, L. Hsu, and D. Kleppner, Phys. Rev. Lett. **62**, 893 (1989).

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