Positive-Energy Structure of the Diamagnetic Rydberg Spectrum

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We report the existence of narrow resonances in the positive-energy region of the diamagnetic spectrum of lithium. The level structure evolves with magnetic field in an orderly fashion. Linewidths corresponding to lifetimes greater than 3000 cyclotron periods have been observed. The regime studied is characterized by chaotic classical motion.

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A Rydberg atom in a strong magnetic field challenges quantum mechanics because it is one of the simplest experimentally realizable quantum systems for which there is no general solution. In addition, it is ideally suited to studying quantum manifestations of classical chaos because it is among the small number of systems in this area that can be studied in detail both experimentally and theoretically. In particular, the Hamiltonian can be varied continuously to carry the system through different regimes of classical behavior. We present here data on the positive-energy spectrum of lithium in magnetic fields of approximately 6 T. The relatively high resolution reveals hitherto unrecognized features, including narrow resonances that extend far above the ionization limit. The lifetimes of these states can be extremely long compared to the time scale for cyclotron motion in these fields. Their structure and their precise ionization mechanism are for the most part a mystery. We believe that an explanation of these long-lived resonances poses a critical test for atomic theory and must be part of any comprehensive explanation of the connection between quantum mechanics and classical chaos.

For a uniform magnetic field in the z direction the system is described in cylindrical coordinates and atomic units by the Hamiltonian

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

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, interactions of the electron and nuclear spins, and effects of the ionic core are neglected. Our experiment employs lithium, rather than hydrogen, for which the Coulomb potential term must be modified in the vicinity of the core. (However, as discussed below, we believe that the effect of the core is unimportant.) Although there is no general solution, considerable progress has been made in understanding the low-energy, low-field regime, where the principal quantum number n is a good quantum number (the "l-mixing" regime), and the adjoining regime at higher field, where n is an approximate quantum number (the "n-mixing" regime).[1] In addition, accurate numerical solutions have been generated above the n-mixing regime at energies up to -20 cm^{-1} in fields up to 6 T.[2] With respect to classical behavior, three separate regimes have been identified.[3] These are characterized, in order of increasing energy, by regular motion, mixed regular and irregular (chaotic) motion, and completely chaotic motion. The results reported here lie entirely in the regime of complete chaos.

The experimental approach is identical to that of a recent study of energy-level statistics in the n-mixing regime.[4] Details of the experiment will be published elsewhere. A highly collimated atomic beam of lithium traveling parallel to a magnetic field is excited to Rydberg states by a two-step process: The $2S\rightarrow3S$ two-photon transition and the $3S\rightarrow$ odd-parity, $m=0$, Rydberg-state transition. The spectral resolution is 10^{-3} cm^{-1} FWHM and the absolute accuracy of the energy measurements is $\pm2\times10^{-3}$ cm^{-1}. The magnetic field is determined[5] with an uncertainty of $\pm5\times10^{-4}$ T.

To set this work into context, we call attention to the elegant measurements by Welge and co-workers at Bielefeld of the absorption spectrum of hydrogen in magnetic fields up to 6 T in the vicinity of the ionization limit.[6] The most prominent characteristic of their data is a regular modulation of the intensity of the spectrum at a frequency of approximately $\frac{3}{2}\omega_c$, where ω_c is the cyclotron frequency of the free electron. This modulation, often called the "quasi-Landau resonance," was discovered many years ago by Garton and Tomkins.[7] However, the data of the Bielefeld group revealed that the quasi-Landau resonance is but one of many periodicities in the absorption spectrum. Furthermore, they found that in many cases the period is equal to the time for the electron to move from the nucleus and return it to by some particular classically allowed path. It should be emphasized that these orbits are *not* periodic in the usual sense: They are unstable and the motion rapidly becomes chaotic.

A semiclassical analysis by Du and Delos[8] has provided a description of this problem. In their analysis, a wave propagates along a classically allowed orbit. The intensity of the resonance is inversely related to the degree of spreading of the wave during a single traversal.

As one expects in a regime of chaos, the waves spread rapidly and contribute little to the intensity after the first traversal. Thus the large-scale modulations in the intensity of the spectrum are associated with wave propagation along closed classical paths, and also with the behavior of the system on a very short time scale.

In the work described here we concentrate on a complementary aspect of the problem: long-term behavior as revealed by high-resolution spectroscopy. Figure 1(a) shows a spectrum extending from $E=0$ to $+30\ \text{cm}^{-1}$ at a field of 6.1 T.[9] (The energy is measured relative to the zero-field ionization limit.) Few details are visible at this scale; indeed the spectrum resembles a picture of noise. However, a Fourier transform of the spectrum has shown pronounced peaks at short times. The transform agrees with the prediction of Du and Delos, very much as in the work on hydrogen. When the spectrum is viewed with higher resolution, however, as in Figs. 1(b) and 1(c), it is discovered to be comprised of a large number of lines, many of which are narrow, and a low but noticeable continuum. Below about $+10\ \text{cm}^{-1}$, some lines are essentially resolution limited, indicating a lifetime longer than

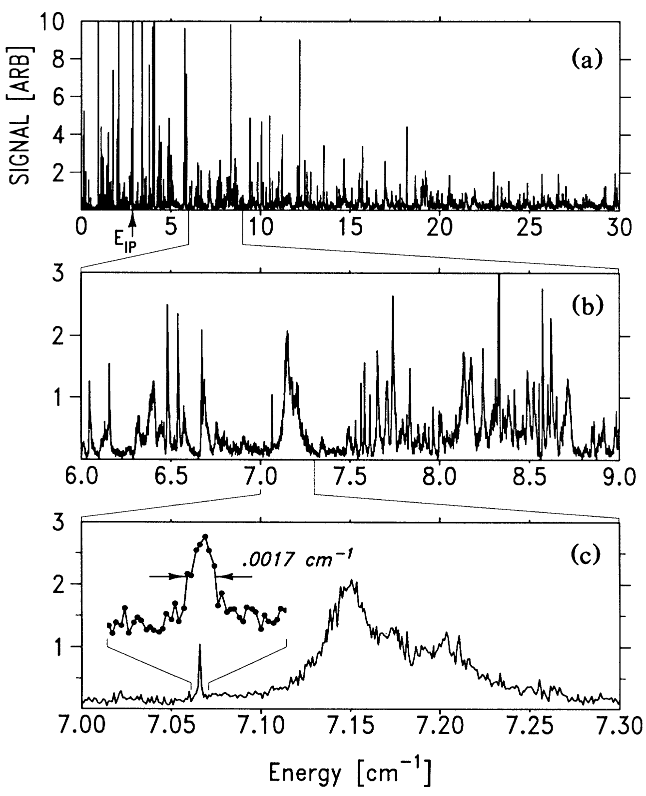

FIG. 1. (a) Lithium diamagnetic spectrum, $m=0$, odd parity, extending from 0 to $+30\ \text{cm}^{-1}$. The signal is proportional to the field-ionization current. Energy is measured relative to the zero-field ionization limit. (b) Tenfold expansion of a portion of (a). (c) Tenfold expansion of a portion of (b). Inset: Tenfold expansion of a narrow line; the dots are data points.

3×10^3 times the cyclotron period. As energy is increased, the lines appear to be broader: The width of the narrowest line observed near $+30\ \text{cm}^{-1}$ is $1.0\times10^{-2}\ \text{cm}^{-1}$. The line shown in the inset of Fig. 1(c) is one of many examples: Its juxtaposition with the broad resonance nearby is typical of the variety of behavior in this region. The occurrence of such long-lived resonances, surprising from a semiclassical point of view, demands explanation for a comprehensive understanding of the system.

Although a quantum solution in the positive-energy regime is lacking, one can make some statements about how the atoms might eventually ionize. The last term in the Hamiltonian of Eq. (1), the diamagnetic interaction, confines the electron in the ρ direction. Hence ionization can only occur in the z direction. The ground-state energy of a free electron in a magnetic field is $E_{\text{IP}}=+\frac{1}{2}\omega_c$: This is the minimum energy for the atom to ionize. In a field of 6.1 T, $E_{\text{IP}}=2.85\ \text{cm}^{-1}$, as indicated in Fig. 1. The narrow states we are considering lie far above E_{IP}. Fano has proposed an approach to the problem based on multichannel quantum-defect theory.[10] A recent adiabatic treatment of the problem by Wang and Greene[11] separates the problem into a rapid transverse motion and a slow longitudinal motion. Using this approach they are able to account for the quasi-Landau resonance. In addition, their model suggests the existence of narrow autoionizing states that decay via coupling with states from different adiabatic potential curves. However, they point out that nonadiabatic effects are large, and further work is needed for a quantitative description.

One might speculate that the sharp spectral features are associated with isolated stable orbits in a chaotic region of phase space about which semiclassical wave functions can be constructed. This immediately raises the question of the stability of the spectrum with respect to the magnetic field. To investigate the stability we have measured a series of spectra at increasing fields in the vicinity of E_{IP}. The results, shown in Fig. 2, reveal that the sharp peaks are not isolated occurrences but the signature of states whose energy increases with magnetic field in an orderly fashion. The overall behavior of the resonances is a monotonic increase in energy with field with slopes between 4 and 10 $\text{cm}^{-1}\text{T}^{-1}$, but the progressions are rich in detail. Some lines appear abruptly and disappear at higher field. Others seem to interact strongly in a manner suggesting anticrossings between levels. Particularly noteworthy is the continuity of the spectrum across the ionization threshold. A useful measure of the extent of the wave functions is the expectation value $\langle\rho^2\rangle$. According to the Feynman-Hellman theorem,[12] the slope of a line in the energy-level map is given by

$$\frac{\partial E}{\partial B}=\left\langle\frac{\partial H}{\partial B}\right\rangle=\frac{1}{4}B\langle\rho^2\rangle+\frac{1}{2}m\,. \qquad (2)$$

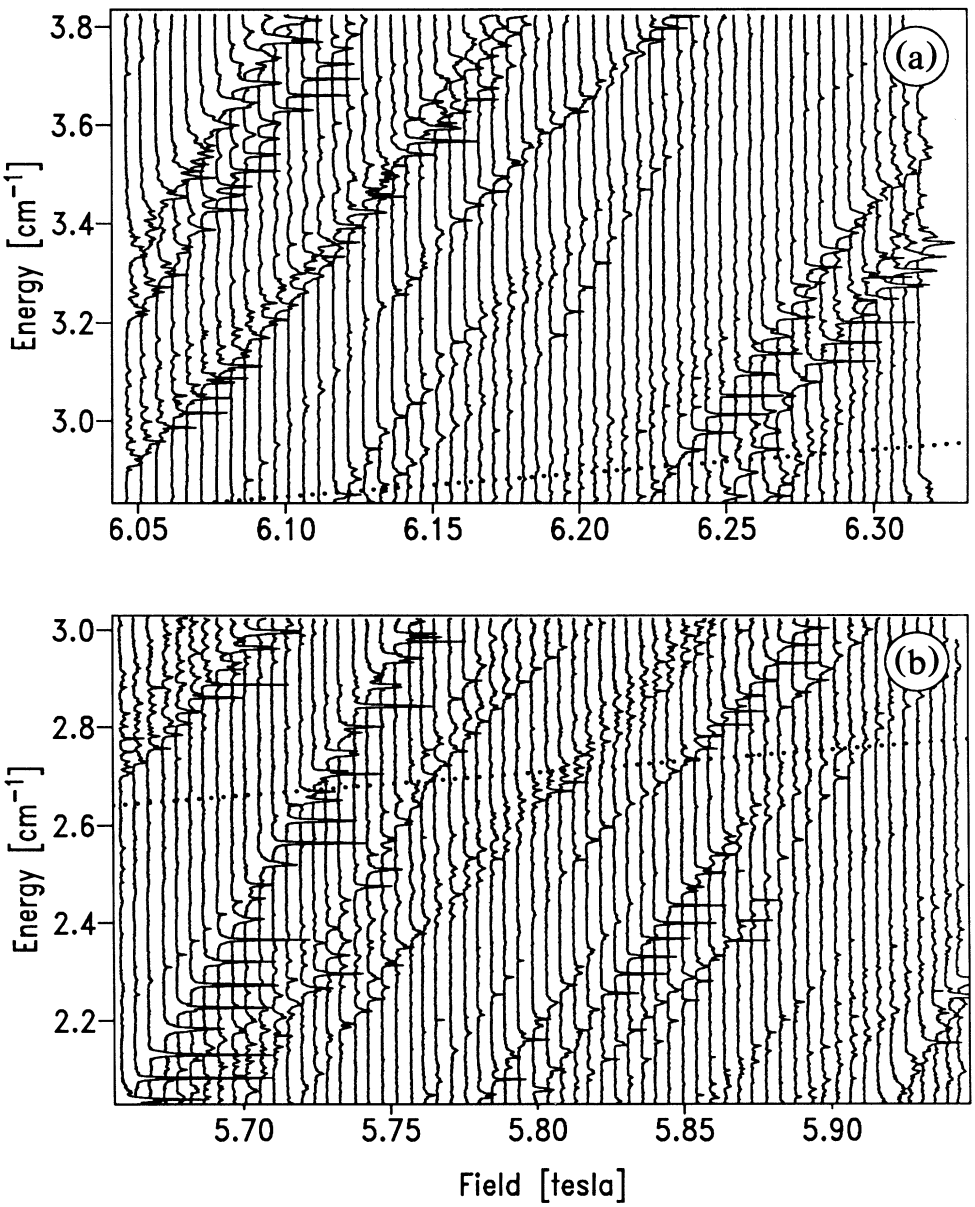

FIG. 2. Energy-level maps for two regions near the ionization threshold. The dotted line indicates the threshold energy $E_{\mathrm{IP}} = + \frac{1}{2}\omega_c$. The maps are created by scanning the laser at a fixed field, and repeating the process at field increments of approximately 4.6×10^{-3} T. The horizontal peaks are field ionization signals. The structure is most easily seen by viewing close to the plane of the page from lower left to upper right.

For $m=0$ states at 6.1 T with a slope of 10 cm^{-1}T^{-1}, this gives $\langle \rho^2 \rangle^{1/2} = 1.3\times10^3 a_0$, where a_0 is the Bohr radius.

In interpreting the data, the fact that Eq. (1) does not include the perturbation of the potential from $-1/r$ at small distances due to the lithium core must be considered. The core perturbation is normally described by quantum-defect theory. For odd-parity states of lithium the largest quantum defect is 0.04. As described elsewhere,[4] the effect of the core in the l- and n-mixing regimes is minor: It is plausible to surmise that the effect of the core is also minor in the positive-energy regime.

Additionally, two experimental problems should be pointed out. The first is the existence of stray electric fields due to surface effects in the interaction chamber. Although these fields cannot be eliminated in our apparatus, they can be measured. A typical value is 0.05(1) V/cm, with the field pointing dominantly along the z axis. Such a field can mix states of opposite parity: Some of the anticrossing features in Fig. 2 are characteristic of this process. Data taken on different days when the stray field differed by 10 mV/cm reveal small but significant differences in some features. Nevertheless, the overall level structure persists. The second problem is that the Rydberg atoms are not detected at the moment they are created. The atoms must drift 2 cm to a region of high electric field where they are field ionized, causing a delay of $\approx 10\ \mu s$. An atom that spontaneously ionizes within this drift region would not be detected if its electron were lost because of the stray electric field. We have no evidence for such a systematic loss of signal.

The development of techniques for precise measurements of the structure of diamagnetic Rydberg atoms opens the way to generating comprehensive energy-level maps in every regime of interest. We hope that these maps will provide useful guides for theoretical interpretation, though the challenge to atomic theory is likely to be formidable. In addition, the existence of long-lived features in a positive-energy regime of chaos poses a serious challenge to our understanding of the connection between chaos and quantum mechanics.

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