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Diamagnetic Structure of Na Rydberg States

Myron L. Zimmerman, Jarbas C. Castro, and Daniel Kleppner

Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 11 January 1978)

We present an experimental survey of a group of Rydberg states of sodium in magnetic fields up to 6 T, where levels from different terms overlap and interact strongly. The observed energy levels and relative line intensities for even-parity $m_l = 1$ terms in the vicinity $n = 28$ are in good agreement with calculations based on a numerical diagonalization using spherical basis states in the range $n = 25$ –31.

The problem of a one-electron atom in an intense magnetic field poses a challenge to atomic physics; in spite of the apparent simplicity of the system, no comprehensive theory exists. (Many theoretical studies have been carried out, however, on problems arising in solid state physics, astrophysics, and atomic spectroscopy.¹) A clear experimental picture of the structure of a simple atom in magnetic fields of increasing strength would be of value both in providing physical insight and in motivating new theoretical attacks.

We present here a step in that direction: a study of the structure of sodium in a field large enough for the diamagnetic interaction to dominate the term separation. We have selected for study states of even parity with azimuthal quantum number $m_l = 1$. As will be discussed below, such states can be described to excellent approximation by an unmodified hydrogenic Hamiltonian.

The eventual goal of this study is to observe a one-electron atom in a field so intense that the Lorentz force on the electron dominates the Cou-

lomb interaction. For an atomic ground state this requires fields in excess of α^{-1} atomic units, or 2×10^5 T (1 T = 10^4 G). Although such fields cannot be achieved in the laboratory, they are of astronomical interest; fields on a neutron star are estimated to reach 10^8 T. The intense field region can be achieved effectively in the laboratory for excitons, hydrogenlike electron-hole systems in semiconductors for which the Coulombic interaction is very weak. Motivated by these problems, the major theoretical effort has been devoted to the ground state of hydrogen and its first few excited states. The intense field region for a hydrogenic atom can also be achieved in the laboratory by employing highly excited states (Rydberg states), because the ratio of diamagnetic energy to binding energy varies as $n^6 B^2$, where n is the principal quantum number.

In a pioneering study with sodium Rydberg states, Jenkins and Segrè² observed the first stages of state mixing due to an off-diagonal term in the diamagnetic Hamiltonian. More recently, Garton and Tomkins³ observed absorption spectra of barium in a field of 3 T. They made use of barium's anomalously large oscillator strength for the principal Rydberg series to study levels up to $n = 80$. A complete elucidation of the level structure of barium is complicated by configuration interaction effects, which are known to play an important role and which make it difficult to identify all the absorption lines. We have chosen to study states in sodium in which many-electron effects are negligible, and to approach the intense-field limit by working our way up from relatively low magnetic fields, hoping to account for all features observed along the way.

The experimental technique is similar to that used in an earlier study of the structure of sodium in a strong electric field.⁴ Figure 1 shows a schematic diagram. An atomic beam is stepwise excited to a Rydberg state by two 5-ns pulsed dye lasers⁵ having linewidth approximately 0.15 cm^{-1} . 1 μs following the laser pulse, a 2-kV/cm electric field is applied to ionize the Rydberg state. The electrons are then accelerated to 10 keV and detected by a surface-barrier diode. The detection scheme is nearly 100% efficient, insensitive to magnetic field, and linear over a wide dynamic range.

The atomic beam travels along the axis of a superconducting solenoid. This geometry essentially eliminates motional Stark fields, a potentially serious problem in experiments employing a gaseous medium. The laser beams are trans-

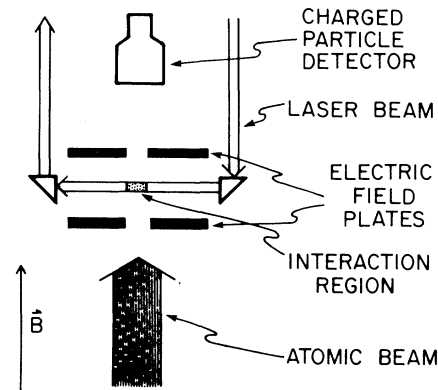


FIG. 1. The apparatus. The region shown is located in the center of the superconducting solenoid. The field plates float at -10 kV with respect to the detector. The pulsed field is applied between the plates.

verse to the atomic beam. One laser saturates the transition $3S_{1/2} - 3P_{3/2}$ at 589 nm; this laser is tuned with the magnetic field to follow the transition $m_j = \frac{1}{2} \rightarrow m_j = \frac{3}{2}$. The second laser, polarized parallel to the field, induces transitions from the level $3P_{3/2}$, $m_j = \frac{3}{2}$ to even-parity $m_j = \frac{3}{2}$ Rydberg states in the vicinity of $n = 28$. Because the laser pulse is short compared to the inverse of the fine-structure frequency of the Rydberg states, m_l (a good quantum number in the intermediate $3P_{3/2}$ "stretched" state) is conserved. The transition satisfies $\Delta m_l = 0$ and populates final states with $m_l = 1$. The linear Zeeman effect is absent. (Even-parity $m_l = 0$ states can also be populated with our pumping scheme, but, because of spin-orbit coupling, always with an admixture of $m_l = 1$ state. To simplify the presentation, only the $m_l = 1$ results are presented here.)

Data are shown in Fig. 2(a). Each frequency scan at a fixed field represents 800 points, with six laser pulses per point.

The energy levels shown in Figs. 2(b) and 2(c), and overlayed on the data in 2(a), were calculated by numerically diagonalizing the Hamiltonian matrix¹

$$\mathcal{H} = \frac{1}{2}(\mathbf{p} + \alpha \mathbf{A})^2 + V(r)$$

(atomic units). The potential $V(r)$ is taken as Coulombic outside of the core, and core effects are taken into account by a quantum-defect treatment. Fine structure is ignored, and linear Zeeman terms, which make no contribution to the spectrum because of the selection rules, are dropped. The remaining field-dependent contri-

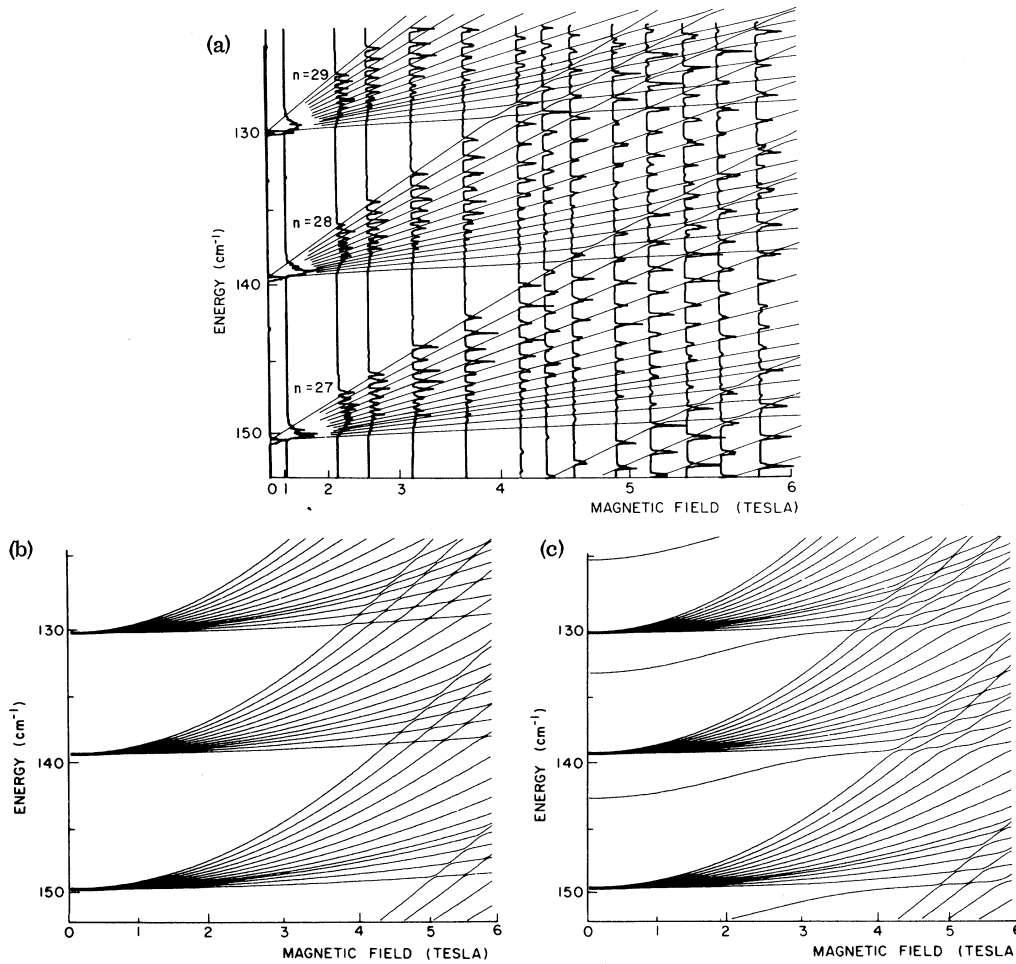


FIG. 2. Diamagnetic structure of Na. (a) Experimental excitation curves for even-parity levels, $m_l = 1$, $m_s = \frac{1}{2}$, in the vicinity of $n = 28$. A tunable laser was scanned across the energy range displayed. The zero of energy is the ionization limit. Signals generated by ionizing the excited atoms appear as horizontal peaks. The horizontal scale is quadratic in field. Calculated levels are overlaid in light lines. Some discrepancies are present due to nonlinearity of the lasers. (b) Calculated excitation curves, displayed linearly in field. (c) Same as (b), but for even-parity $m_l = 0$ states. Note the large effect on anticrossings due to the presence of the nondegenerate s states.

bution to the Hamiltonian is the diamagnetic term

$$\begin{aligned} \frac{1}{2} \alpha^2 A^2 &= \frac{1}{8} \alpha^2 B^2 r^2 \sin^2 \theta \\ &= \frac{1}{12} \alpha^2 B^2 r^2 [1 + (4\pi/5)^{1/2} Y_{20}], \end{aligned}$$

which has matrix elements connecting states with the same azimuthal quantum number and the same parity. It should be pointed out that the diamagnetic interaction is physically distinct from the quadratic Zeeman effect which arises from breakdown of $\vec{L} \cdot \vec{S}$ coupling (the Paschen-Back effect). Furthermore, in contrast to previous studies in which nonhydrogenic effects split the l degeneracy at low field, in the case of a Coulomb potential all states of identical parity and identi-

cal m_l are mixed by a vanishingly small field. Thus " l -mixing" effects are intrinsic to the system, though n remains a good quantum number at low fields.

A basis of zero-field spherical states, extending over all even-parity $m_l = 1$ states in the range $n = 25-31$, was used in the diagonalization. Radial matrix elements were generated by using the Coulomb approximation and numerically integrating the radial equation. Quantum defects were calculated from spectroscopic data and the core polarization.⁶ The energy eigenvalues were obtained as a function of magnetic field at 0.01-T increments, and interpolated for intermediate field values. Nonhydrogenic effects for the $m_l = 1$

state Fig. 2(b), are present, but are too small to be visible in the drawing. They are quite conspicuous in the results for $m_l=0$, Fig. 2(c), due to the nonhydrogenic s state, though the calculation may have exaggerated some of the anticrossings. As an indication of the breakdown of n as a good quantum number, the upper components of the $m_l=1$, $n=28$ manifold at 3.5 T have a 20% contribution (sum of amplitude squared) from other terms.

The chief sources of discrepancy in Fig. 2(a), aside from the laser resolution, are a slight nonlinearity in the frequency scan, apparent in the systematic offset of the levels for the $n=29$ manifold, and uncertainty in the field calibration. Because the level structure provides a more sensitive calibration of the field than the magnet-current monitor, the final field values were obtained by fitting the data at three field values. (The fits all agreed with the field calibration within the 5% uncertainty of the current monitor.) We feel that theoretical and experimental energy levels agree within experimental error.

We have also considered the line intensities. The eigenvectors were calculated from the diagonalization procedure described above, and used to obtain relative line intensities (approximately the square of the amplitude of the d -state character). At 4 T for $n=28$ there is a predicted intensity ratio of ~ 9 between the high- and low-energy components. Although laser intensity fluctuations make intensity measurements somewhat unreliable, the data are consistent with this ratio.

To investigate the sensitivity of the energy-level structure to electron-core interaction, we have carried out the identical calculations for hydrogen. The level plot for the $m_l=1$ even-parity system is essentially indistinguishable from Fig. 2(b). The $m_l=0$ level structure of hydrogen, however, is qualitatively different from Fig. 2(c). The reason is that the largest quantum defect for the former system is 0.014, due to the d state, while for the latter it is 1.35, due to the s state. As in the case of Stark structure, the diamagnetic level structure is very sensitive to large quantum defects but generally insensitive to quantum defects $\ll 1$.

The numerical calculations suggest that as the system becomes hydrogenic (i.e., the quantum

defects all vanish) the levels from different terms may actually cross, analogously to level-crossing behavior in an electric field.⁷ This would imply that a dynamical symmetry exists, although it is probably not of a simple nature as in the Stark problem where the symmetry is manifest in the separation of the Hamiltonian in parabolic coordinates.

Although we have been able to account for the observed energy levels by a straightforward diagonalization procedure, it should be pointed out that the method is at its limit of usefulness and that new theoretical approaches are badly needed. The experimental technique can be extended to somewhat higher fields, and, with better laser resolution, to much higher term values. Thus one can look forward to detailed data on hydrogenic systems for both the intermediate- and intense-field limits.

We would like to thank Dr. Michael G. Littman for his valuable assistance with the lasers, Dr. Aram Mooradian for the use of the magnet, and the Conselho Nacional de Pesquisa (Brazil) for partial support of J. C. Castro. This work was sponsored by the National Science Foundation.

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