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AVOIDED CROSSINGS AND AUTOIONIZATION WIDTHS OF HYDROGEN AND ALKALI METALS IN MAGNETIC FIELDS

M.L. Zimmerman, M.M. Kash and G.R. Welch

Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, U.S.A.

ABSTRACT
Anticrossing sizes and autoionization widths provide sensitive tests of theories on hydrogen in magnetic fields. The various contributions to these sizes and widths are discussed, as well as calculational methods. An experimental method is described for studying these effects by cw laser spectroscopy on lithium.

INTRODUCTION
The spectra of highly excited atoms in magnetic fields show a remarkable simplicity which was not expected from early theoretical considerations. Garton and Tomkins initially observed "quasi-Landau" resonances as field dependent periodic modulations of the oscillator strength with energy near the ionization limit. These observations were subsequently explained and calculated by various authors using several methods. The discovery of the "quasi-Landau" levels stimulated much interest in the structure of atoms in strong fields.

Another aspect of the simplicity, perhaps even more profound, is the apparent crossing of levels in hydrogen. This suggestion of an unexpected symmetry surfaced during numerical calculations of the spectra of alkali metals and hydrogen. Figure 1 shows the energy levels and several anticrossings for hydrogen. The anticrossings between adjacent n-manifolds become small quite rapidly as n increases. Although the approximate crossings are from computational results, interestingly, the observation is empirical. Figures 2 and 3 show "true" experimental results for sodium which demonstrate the accuracy of...
the calculations and also suggest level crossings. Several recent calculations\(^5\), spurred in part by the level crossing observation, have made progress, especially in the low field region (when \( n \) mixing is small) and estimate both energies and wavefunctions.

![Energy of hydrogen as a function of magnetic field](image)

**FIG. 1a.** Energy of hydrogen as a function of magnetic field (plotted on a squared scale) for several low-lying states. The energy is measured from the zero-field ionization limit. The level repulsion of the circled "crossing" is about 8 cm\(^{-1}\).

**PROPOSED METHOD**

Although there are no known separations of the hamiltonian for hydrogen in a magnetic field, an approximate symmetry suggests the following form:

\[
H = \frac{1}{2} \mathbf{p}^2 - \frac{1}{r} - \frac{1}{8} \alpha^2 \mathbf{r} \cdot \mathbf{p}^2 = H_S + H_{SB}
\]

(1)

where \( H_S \) is the part of the hamiltonian exhibiting the symmetry and \( H_{SB} \) is the symmetry breaking part of the hamiltonian. \( H_S \) becomes the total hamiltonian at zero field and the spectra of \( H_S \) show level crossings between levels from different principal quantum numbers. Many of the properties of \( H_S \) are analogous
FIG. 1b. Energy of hydrogen as a function of magnetic field (plotted on a squared scale) for several higher excited states.

FIG. 2. Diamagnetic structure of Na. Experimental excitation curves for even-parity levels, $m_s = 1$, $m_l = -1$, in the vicinity of $n = 26$. Calculated levels are overlaid in light lines.
to the Hamiltonian of hydrogen in an electric field. However in the electric field case, the separation and symmetry are well known and can be used to help solve the problem. Anticrossings of the energy levels of $H$ which cross for $\mathcal{H}_S$ are due to the contribution from $\mathcal{H}_{SB}$. Figure 4 graphically shows the anticrossing size of the lowest field "level crossing" between adjacent $n$ manifolds as a function of $n$. Since this size decreases very rapidly as $n$ increases, then $\mathcal{H}_{SB} \ll \mathcal{H}_S$ for many regions of interest and can be ignored or treated perturbatively.

![Graph showing anticrossing size of the lowest field level crossing between adjacent n manifolds](image)

**FIG. 3.** Experimental excitation curves for even-parity, $m_L = -2$ energy levels of sodium plotted as a function of magnetic field (plotted on a squared scale). Signals generated by ionizing the excited atoms appear as horizontal peaks. The level structure is most clearly seen by sighting along the drawn lines with the eye close to the plane of the paper. Solid lines indicate the evolution of the highest level for several $n$ manifolds. The dashed and dot-dashed lines show the second- and third-highest levels of the $n = 40$ manifold, respectively.

The preceding method needs to be tested. The numerical calculation which leads to the observation of crossings cannot reliably be applied to very high $n$ or higher magnetic field anticrossings. In particular, it cannot determine the
magnitude of $H_{SB}$ in many regions of interest. However, the anticrossing size can be measured by experiments using high resolution cw dye lasers. Also, the autoionization widths of levels above the ionization limit are influenced by $H_{SB}$. In this case many states of $H_S$ are bound, even above the ionization limit. An analogous situation takes place for hydrogen in an electric field\(^6\). $H_{SB}$ then couples these states to continuum states of $H_S$, leading to autoionizing states. A cw laser experiment can be used to measure the level widths. Thus the same experiment can determine $H_{SB}$ over a wide energy and magnetic field range.

**FIG. 4.** Anticrossing size as a function of $\bar{n}$, the arithmetic mean of the principal quantum numbers of the adjacent manifolds. The solid line depicts the anticrossing size between the lowest-energy state of the $n+1$ manifold and the highest-energy state of the $n$ manifold. The dashed line shows the anticrossing size between the lowest energy state of the $n+1$ manifold and the "middle" energy state of the $n$ manifold. (The fluctuation of the points about the lower end of this line reflects the logarithmic ordinate and the problem of defining the "middle" of a manifold.)
Effects of the Core for Alkali Metal Atoms

Alkali metal atoms are convenient to use in laser spectroscopy experiments. However, the core electrons of these atoms have a short range interaction which contributes both to the anticrossing sizes and autoionization widths. These core contributions must be understood and calculated before experimental measurements of alkali metal atoms can be related to the symmetry breaking part of magnetic hamiltonian $H_{SB}$.

The hamiltonian for an alkali metal atom in a magnetic field is written as:

$$H = H_S + H_{SB} + V_{core}$$

where $H_S$ and $H_{SB}$ are the same as defined in Equation 1 and $V_{core}$ is a short range central potential due to the core electrons. Except for $H_{SB}$, Equation 2 is similar to the hamiltonian for alkali metal atoms in electric fields, which is written as:

$$H = H_{hy} + V_{core}$$

Here $H_{hy}$ is the hamiltonian for hydrogen in an electric field and $V_{core}$ is the same term as in Equation 2. However the anticrossings for the electric field hamiltonian of Equation 3 can be calculated very accurately using a perturbation method described by Komarov. It is worthwhile reviewing this method as applied to the electric field case before developing a slightly modified method for the magnetic field case.

Komarov Method for Electric Fields

The method begins by writing the short range central core potential as

$$V_{core} = \Sigma_{l} v_l(r) P_{l}$$

where $P_{l}$ is the projector operator into the $l$ angular momentum subspace for the valence electron. The radial part of the spherical basis wavefunction is expanded as functions of $r$ in the energy $E$ (or $n^{-2}$) about $E=0$. 
The matrix elements of $V_{\text{core}}$ are:

$$
\langle n'\ell' m' | V_{\text{core}} | n\ell m \rangle = \delta_{\ell \ell'} \delta_{mm'} (nn')^{-3/2}
$$

The integrals are replaced by the parameters $-\delta_O^\ell, -\delta_2^\ell/2$ which depend only on $\ell$ and the particular atom. The matrix elements reduce to:

$$
\langle n\ell m | V_{\text{core}} | n'\ell' m' \rangle =
$$

$$
\delta_{\ell \ell'} \delta_{mm'} (nn')^{-3/2} (\delta_O^\ell + (n^{-2} + n'^{-2})\delta_2^\ell/2 + 0(n^{-4}))
$$

These $\delta_s$ can be related to the zero electric field energies by expressing the energy to first order:

$$
E_{n\ell m} = E_n + \langle n\ell m | V_{\text{core}} | n\ell m \rangle
$$

$$
= - \frac{1}{2} n^{-2} - n^{-3} (\delta_O^\ell + n^{-2}\delta_2^\ell + 0(n^{-4}))
$$

Thus the $\delta$ parameters can be determined by fitting them to zero field spectroscopic data. The parameter $\delta_O^\ell$ can be identified as the normal energy independent quantum defect.

In hydrogen the well known parabolic separation allows levels from different $n$'s to cross in an electric field. However in the alkali metal atoms, the core destroys the parabolic separation and the level crossings become anticrossings. Degenerate perturbation theory predicts that the anticrossing size is twice the matrix element coupling the two crossing states:

$$
\Delta E = 2|\langle n\ell m | F | V_{\text{core}} | n'k'm' | F \rangle|
$$
where \( k \) is the parabolic quantum number \( (k = n_1 - n_2) \). The correct unperturbed wavefunctions are analytically known at zero field (the parabolic states), and are only slightly modified by the field, particularly near the core where the coupling is strongest. Thus to an excellent approximation, the states in the field can be replaced by the (zero field) parabolic states. In order to calculate the matrix element in equation 9 the parabolic states must be mapped to spherical states, because \( V_{\text{core}} \) is characterized by parameters of the spherical states, that is the quantum defects. This mapping is a simple transformation \(^{10}\) given by:

\[
|nkm\rangle = \sum_{l=|m|}^{n-1} \frac{1}{\sqrt{\Sigma}} <\frac{n-1}{2}, \frac{n-1}{2}, \frac{m+k}{2}, \frac{m-k}{2}|\ell m\rangle |n\ell m\rangle
\]  

(10)

where \(<|\rangle \) are Clebsch-Gordan coefficients. The anticrossing size given by equation 9 can be rewritten as:

\[
\Delta E_{nkm; n'k'm} = (nn')^{-3/2} \sum_{l=|m|}^{n-1} \frac{1}{\sqrt{\Sigma}} <\frac{n-1}{2}, \frac{n-1}{2}, \frac{m+k}{2}, \frac{m-k}{2}|\ell m\rangle \times
\]

\[
<\frac{n'-1}{2}, \frac{n'-1}{2}, \frac{m+k'}{2}, \frac{m-k'}{2}|\ell m\rangle \delta_{\ell}^{\ell} + (n^2+n'-2)\delta_k^k / 2
\]  

(11)

Figure 5 compares several anticrossings sizes in lithium for experimental results, numerical diagonalizations, and equation 11. (The states are labeled by parabolic quantum numbers.) These results are quite close and indicate that Komarov's approach is valid, at least for small quantum defects.

A modified approach for magnetic field

The \( V_{\text{core}} \) matrix elements can also be calculated for magnetic field states, however there is no known simple mapping of magnetic field states to spherical states. The magnetic field states, at low fields where \( n \) mixing is negligible, are numerically generated by diagonalizing the magnetic field Hamiltonian including all spherical states of a given \( n, m \) and parity. These states are analogous to the zero electric field parabolic states, except that instead of Clebsch-Gordan coefficients relating the parabolic to spherical states, the coefficients of the eigenvectors from the diagonalization relate the magnetic to spherical states.
The preceeding calculation can be compared with the anticrossing sizes from "large" basis numerical diagonalizations including states from many n's. Since the diagonalization results are most accurate at lower fields, the comparison is performed on the first anticrossing between adjacent n's. Also, a simple fictitious alkali metal atom, testium, is introduced for the purpose of making the comparison. Only the "s" states of testium have nonzero quantum defects ($\delta_0 \neq 0$) and thus the anticrossing size scales linearly with the quantum defect, as long as the two state approximation of degenerate perturbation theory is appropriate. Figure 6 shows the anticrossing size multiplied by $n^2(n+1)^2/\delta_0$ as a function of $n$ for the first anticrossing of testium in a magnetic field. The first anticrossing of testium in an electric field is also shown for reference and is constant at 2. (The magnitude of the Clebsch-Gordan coefficient for testium in an electric field is $n^{-1/2}$.) The "large" basis diagonalization results for the magnetic field give a constant of roughly 5, while the perturbation method gives a value which increases from about 4 to 6 as $n$ increases from 10 to 40. The difference could be due to errors caused by a truncated basis set used in the "large" basis numerical diagonalization, or errors in the eigenvector coefficients caused by neglecting $n$-mixing in the perturbation method. However, these effects do not seem large enough to account for the difference. Effort continues in resolving this discrepancy.
FIG. 6. Normalized anticrossing size as a function of \( \bar{n} \), the arithmetic mean of the principal quantum numbers of the adjacent manifolds. The particular anticrossings shown are between the lowest energy state of the \( n+1 \) manifold and the highest energy state of the \( n \) manifold. The solid line displays the perturbation calculation, and the X's show the results of large basis diagonalization, both for testium in a magnetic field. The dashed line is the perturbation result for testium in an electric field.

\( V_{\text{core}} \) also couples bound states, which are energetically above the ionization limit, to unbound or continuum states. The width of the autoionizing levels is given by the matrix element of \( V_{\text{core}} \) between the degenerate bound and continuum states. The bound states are well characterized by the preceding perturbative anticrossing analysis. However, the continuum states need to be generated and projected onto a spherical basis before any widths caused by core effects can be predicted. 11

Experiments with lithium

Since the Rydberg levels of hydrogen are not easily accessible with lasers, lithium is the most desirable atom to use in experiments checking magnetic field symmetry breaking. Lithium has relatively small quantum defects: .40 for the "s" states and .05 for the "p" states. Thus lithium closely
approximates hydrogen for $|m| \geq 1$. The table in figure 7 lists anticrossing sizes calculated using the preceding perturbative analysis for $|m| = 1$, odd parity states of lithium around $n = 40$. These sizes of roughly $0.01 \text{ cm}^{-1}$ or $300 \text{ MHz}$ are easily resolvable with modestly high resolution cw dye lasers. Thus the experiment is to measure various anticrossings in magnetic fields from 1 to 10 Tesla at energies between $70 \text{ cm}^{-1}$ and the ionization limit. Autoionization widths above the ionization limit will also be measured for fields in the 1 to 10 Tesla range.

<table>
<thead>
<tr>
<th>States $(n \ k \ m) \times (n' \ k' \ m)$</th>
<th>Anticrossing Size (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(40,1,1) \times (41,39,1)$</td>
<td>0.0070</td>
</tr>
<tr>
<td>$(40,1,1) \times (41,39,1)$</td>
<td>0.0094</td>
</tr>
<tr>
<td>$(40,1,1) \times (42,39,1)$</td>
<td>0.0109</td>
</tr>
<tr>
<td>$(40,1,1) \times (42,39,1)$</td>
<td>0.0066</td>
</tr>
<tr>
<td>$(40,1,1) \times (42,39,1)$</td>
<td>0.0089</td>
</tr>
<tr>
<td>$(40,1,1) \times (43,41,1)$</td>
<td>0.0033</td>
</tr>
<tr>
<td>$(40,1,1) \times (43,41,1)$</td>
<td>0.0062</td>
</tr>
<tr>
<td>$(40,1,1) \times (43,39,1)$</td>
<td>0.0084</td>
</tr>
<tr>
<td>$(40,1,1) \times (43,39,1)$</td>
<td>0.0097</td>
</tr>
<tr>
<td>$(40,3,1) \times (41,39,1)$</td>
<td>0.0046</td>
</tr>
<tr>
<td>$(40,3,1) \times (41,39,1)$</td>
<td>0.0063</td>
</tr>
<tr>
<td>$(40,3,1) \times (41,39,1)$</td>
<td>0.0072</td>
</tr>
<tr>
<td>$(40,3,1) \times (42,41,1)$</td>
<td>0.0044</td>
</tr>
<tr>
<td>$(40,3,1) \times (42,39,1)$</td>
<td>0.0059</td>
</tr>
<tr>
<td>$(40,3,1) \times (42,39,1)$</td>
<td>0.0068</td>
</tr>
<tr>
<td>$(40,3,1) \times (43,41,1)$</td>
<td>0.0041</td>
</tr>
<tr>
<td>$(40,3,1) \times (43,39,1)$</td>
<td>0.0036</td>
</tr>
<tr>
<td>$(40,3,1) \times (43,39,1)$</td>
<td>0.0065</td>
</tr>
</tbody>
</table>

FIG. 7. Table of anticrossing sizes for lithium in a magnetic field, obtained by the perturbation calculation. Only $m = 1$, odd parity states are shown.

Figure 8 shows the laser excitation scheme for lithium. The superconducting magnet is set to the desired field value and the cw laser for the final excitation step is scanned over the energy of interest. The field value is then reset and the laser again scanned. This process is repeated until the anticrossing is mapped out. An autoionization width is somewhat easier to measure as only one magnetic field and laser scan is required. The resolution of the experiment is limited by the intrinsic linewidth of the 3s state to about 8 MHz. If the final laser is linearly polarized perpendicular to the magnetic field, both the $m = 1$ and $m = -1$ final states are excited. Thus the
experimentally observed linear Zeeman splitting of the final state shows as a doubling of the spectra with a spacing of $\alpha E$.

Final Step $\Delta m_L = 0$

Final Step $\Delta m_L = \pm 1$

FIG. 8. Proposed excitation scheme for populating the Rydberg p-series of lithium. The first step is a two-photon excitation of the 3s state. The second step is a resonant excitation of the Rydberg state. $\Delta m_L$ is determined by the polarization of the final laser relative to the magnetic field.

The experimental apparatus consists of a superconducting solenoid magnetic, an effusive atomic beam shown in figure 9a and an interaction region shown in figure 9b. The atomic beam is parallel to the magnetic field to reduce motional electric fields. The lasers are perpendicular to the magnetic field and intersect the atomic beam to give a several cubic millimeter interaction volume. Either the excited atoms are allowed to drift into an electric field region or a pulsed electric field is applied. In any case, the electric field ionizes the excited atoms and the resulting electrons are accelerated to about 15 kev and detected by a surface barrier diode. The charge on the diode is monitored as a function of laser frequency by a laboratory computer and the spectra are recorded.
Conclusions

Results from these experiments using a cw dye laser are well suited to testing calculations of anticrossings and autoionization line shapes of alkalis and hydrogen in magnetic fields. Flexibility in selecting $n$ or energy, $m$ or the quantum defects, and particular levels provides a wide range of conditions
for tests. Although the ultimate aim is to study the approximate symmetry of hydrogen in a magnetic field, the core effects must first be understood. The core effects are important and interesting on their own, and in fact will help in the understanding of hydrogen's approximate symmetry. Several calculational methods for core effects appear to predict realistic results, although improvements are needed.

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References


11. An alternate treatment of this phenomenon is given by D.A. Harmin, to be published.