Quasi-Landau Modulations in Nonhydrogenic Systems in a Magnetic Field

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The bound-state photoabsorption spectra for Rydberg states of lithium and strontium in a magnetic field are evaluated and compared with that of hydrogen. The line positions and oscillator strengths depend strongly on the Rydberg electron's interaction with the ionic core. The positions of the peaks in the Fourier transform of these spectra, for short times, are insensitive to the strength of the core interaction but the relative intensities of the peaks do depend on this interaction. The energy spacing of $\sim 0.64\omega_c$ is seen in the Fourier transform of all of the spectra.

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The study of a Rydberg atom in a magnetic field has recently begun to address the question of the role of classical chaos and semiclassical quantization in a quantum system. Classically, for laboratory strength fields, the trajectories generated by the Hamiltonian for this system exhibit a transition from regular quasiperiodic motion to irregular or chaotic motion in the vicinity of the ionization threshold.[1] The probability distribution for nearest-neighbor energy-level spacings shows a corresponding transition from a Poissonian distribution in the regular or perturbative region of the spectrum to a Wigner distribution, derived from random-matrix theory, in the irregular or so-called "strong mixing region" near the ionization threshold.[2] This transition can be viewed as the breakdown of all the approximate global constants of the motion[3] present in the perturbative region of the spectrum.

The moments of the theoretical probability distribution, in the strong-mixing region, depart from the predictions of random-matrix theory, demonstrating the existence of long-range correlations or fluctuations compared to the average energy-level spacing.[3] The fluctuations are particular to the diamagnetic system and are not universal in character. Semiclassical theories have aimed at evaluating these fluctuations by calculating the periods and Lyapunov exponents of all of the shortest unstable periodic orbits in the chaotic region.[4] Quantal calculations have used, on the other hand, large basis sets to solve the Schrödinger equation numerically for an atom in a magnetic field.[5,6] This has led to a detailed line-by-line comparison between the theoretical and experimental photoabsorption spectra of hydrogen to within 5 cm^{-1} of the ionization threshold.[7] The recent availability of high-resolution experimental photoabsorption spectra for hydrogen,[8] lithium,[9,10] and barium[11] in a magnetic field now permits a detailed comparison to be made between theory and experiment for these atoms and allows an evaluation of the various semiclassical[4,12] and quantal approaches to this problem.[5,6]

I present, in this Letter, the first theoretical calculation of the photoabsorption spectrum of lithium in a magnetic field in the strong-mixing region near the ionization threshold. This has been the subject of recent experimental investigations.[9,10] In addition, I address the more general question of how the Rydberg electron's interaction with the ionic core affects the spectrum of an atom in a magnetic field in this region where the corresponding classical solutions become chaotic.

The Hamiltonian for the hydrogen atom in a magnetic field, in atomic units, is

$$H = -\tfrac{1}{2}\nabla^2 - 1/r + \beta L_z + \tfrac{1}{2}\beta^2 r^2 \sin\theta\,, \qquad (1)$$

where $\beta = \omega_c/2$, and ω_c is the cyclotron frequency. The magnetic field vector is chosen to be along the z axis. The z component of the orbital angular momentum, L_z, is a conserved quantity since the potential does not depend on the coordinate ϕ. The eigenstates of (1) can be labeled with a fixed value of the magnetic quantum number m. In addition, the potential is symmetric under the transformation $z \rightarrow -z$ so that the eigenstates can be further classified according to their parity, π_z, which can be even or odd under reflection through the $z=0$ plane. Therefore, the eigenstates can be studied within a fixed subspace defined by definite values of m and parity π_z. The conservation of m reduces the system to a two-dimensional nonseparable Schrödinger equation. This equation has been solved and the bound-state photoabsorption spectrum of hydrogen evaluated, by expanding the wave function in a basis set[5] and diagonalizing the Hamiltonian matrix given by (1). These basis-set methods were extended in a general theory of nonhydrogenic systems[6] which used quantum-defect theory[13] to describe the interaction of the Rydberg electron with the ionic core and the R-matrix method to solve the Schrödinger equation, with the Hamiltonian given in (1), over the semi-infinite region outside the core. The quantum defects, μ_l, of the lth partial wave, which represent the effect of the core, are generally well known from the field-free spectra or from *ab initio* calculations. The theory was first used successfully[6] to reproduce the experimental absorption spectrum[14] of strontium and barium in a field of 4.7 T in the inter-l and inter-n mixing regions.

The theoretical excitation scheme used in the present study is chosen to coincide with that used in the recent experimental measurements on Li in a magnetic field.[9] The experiment was performed in a magnetic field of 6.1 T and involved initially preparing the lithium atom in its $3s$ state through a two-photon transition from the Li($2s$) ground state. The Li($3s$) was then excited by linearly polarized light into an odd-parity Rydberg or continuum state with $m=0$. The theoretical photoabsorption spectra presented here for H, Li, and Sr, respectively, are also calculated in a field of 6.1 T. The initial states in the photoabsorption process are the H($3s$), Li($3s$), and Sr($5s^2\,{}^1S^3$), $m=0$ states and the final states are the $m=0$, odd-parity Rydberg states which can be reached by linearly polarized light. (The Sturmian functions were taken as a radial basis set and the largest dimension of the matrices used was 1600. The calculation of all the eigenvalues and eigenvectors for this matrix took about 1.5 h on a Cray X-MP/48 computer.) The structure of the spectrum, over the narrow energy ranges considered here, can be independent of the initial state chosen for a given atom and a particular final-state symmetry. For example, one gets the same form of spectrum by photoabsorption from the H($2s$) initial state as that from the H($3s$) state. The two spectra only differ by an overall constant since the variation of the size of the dipole matrix element with energy is given by the p component of the final-state wave function.[6] The structure of the spectrum is determined by the final state only. Therefore, the differences documented below between the photoabsorption spectra of H, Li, and Sr are due to the Rydberg electron's interaction with the ionic core in the presence of a magnetic field.

I compare in Fig. 1 the photoabsorption spectra of H, Li, and Sr in a field of 6.1 T as a function of the energy of the Rydberg electron in the interval $[-260,-80]$ cm^{-1}. The oscillator strengths for H are absolute but for Li and Sr they are normalized relative to the field-free oscillator-strength distribution (df/dE) of hydrogen. The absolute oscillator strengths are obtained by just multiplying these spectra by a constant that can be obtained from their field-free spectra.[6] The interval shown contains the perturbative inter-l mixing region, where the different n manifolds do not mix, and also the inter-n mixing region of the spectrum. The effect of the core in these regions of the spectrum has been well documented before.[6,10,15] The lowest orbital angular momenta are the only ones which penetrate the ionic core and that have accordingly significant quantum defects. The calculated spectra are odd in parity, π_z, and so the p and f waves are the lowest allowed values of the orbital angular momenta with nonzero quantum defects. Lithium is quasihydrogenic in that the p quantum defect is very small, $\mu_p=0.053$ $(\mu_f=0.0)$.[10] The overall appearance of the spectrum in Fig. 1(b) is similar to that of hydrogen in Fig. 1(a) but the quantum defect leads to most of the oscillator strength, in the inter-l mixing region, being taken up by the np states, resulting in a larger line strength, compared to hydrogen, for the lowest-energy line in a particular n manifold.[10] The Sr spectrum in Fig. 1(c) is more typical of nonhydrogenic systems in that the quantum defect of the p wave in this region is large, $\mu_p \approx 0.72$ $(\mu_f \approx 0.08)$.[6,16] This breaks the degeneracy in the orbital angular momentum which is present in hydrogen in zero magnetic field and results in almost all of the oscillator strength being taken up by the np Rydberg states in the inter-l mixing region. The oscillator strength is redistributed when the n manifolds in Fig.

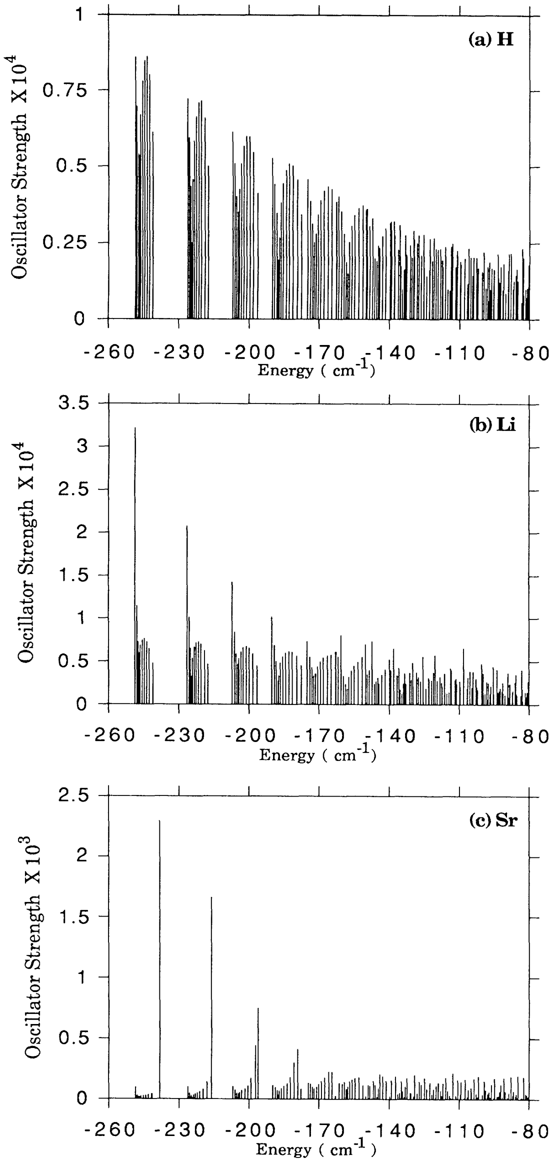

FIG. 1. The oscillator strength as a function of the energy in wave numbers in a magnetic field of 6.1 T for the $m=0$, odd-parity Rydberg states of (a) hydrogen, (b) lithium, and (c) strontium.

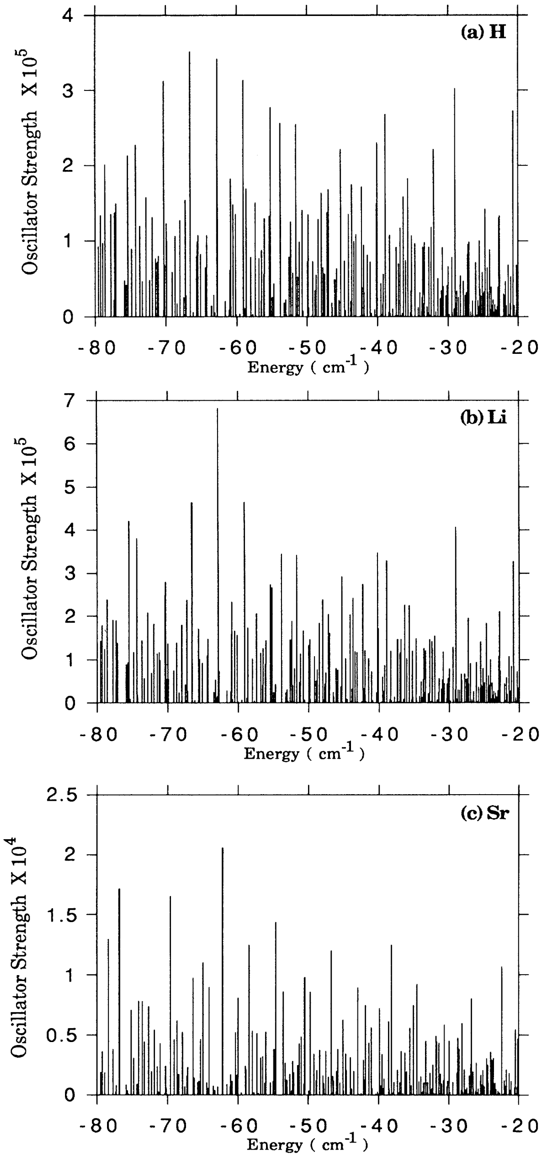

FIG. 2. The same as Fig. 1 but at an energy closer to the ionization threshold.

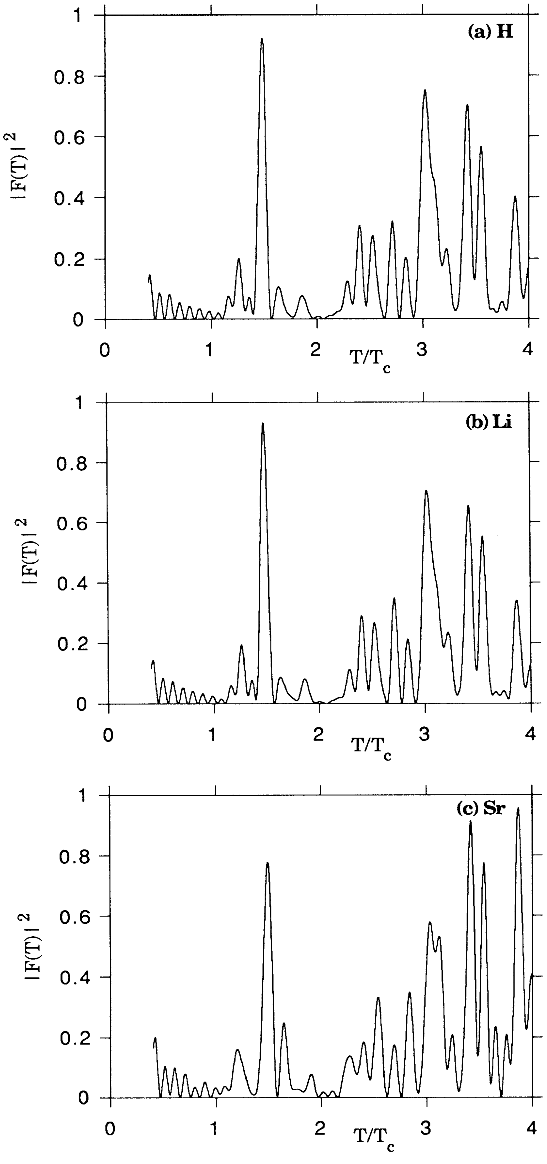

FIG. 3. The square modulus of the discrete Fourier transform of the spectra shown in Fig. 2, in arbitrary units, as a function of time in units of the cyclotron period for (a) hydrogen, (b) lithium, and (c) strontium.

1(c) begin to mix.

The spectra in the energy range $[-80, -20]$ cm^{-1} are shown in Fig. 2 where the corresponding classical problem becomes chaotic.[1] The probability distribution for nearest-neighbor energy-level spacings has a Wigner-type distribution in this region.[2] The spectra in Fig. 2 are extremely complex; nevertheless, a line-by-line comparison has been made between theory and experiment, for the photoabsorption spectrum of hydrogen in a magnetic field, in this energy region.[7] This type of comparison is now also possible for any nonhydrogenic system using the theory outlined earlier. Even though the quantum defect for the p states of Li is very small, $\mu_p = 0.053$, there are strong perturbations of the absorption spectrum in this energy region as can be seen by comparing Figs. 2(a) and 2(b). It is, however, possible to identify groups of lines in Fig. 2(b) which are not significantly perturbed by the presence of the ionic core, so that the perturbations, for Li, tend to be localized. This is not the case for Sr, in Fig. 2(c), where there is no obvious correspondence between this spectrum and that of hydrogen in Fig. 2(a).

The Fourier transform of the photoabsorption spectra has proved very useful in disentangling such complex spectra[8] as those shown in Fig. 2, since the transform lays emphasis on the short-time behavior of the system or correspondingly the modulations in the spectrum which occur over large energy intervals compared to the mean energy-level spacing. These are the modulations or fluctuations mentioned in the introductory paragraphs. The transform of the experimental photoabsorption and photoionization spectra of hydrogen in a magnetic field revealed new periodicities[8] or "quasi-Landau modulations" of $0.64\omega_c$, $0.39\omega_c$, $0.28\omega_c$, etc. These frequencies were related to the recurrence times of unstable periodic orbits in the classically chaotic region.[8,17] The discrete Fourier transform of each of the spectra in Fig. 2 is shown in Fig. 3 as a function of time. The transforms of all three spectra are very similar in that the number and the position of the peaks in each of the spectra are the same. However, there is a difference in the relative intensity of the peaks for Sr compared to those in H. Thus the core does not appear to affect the position of the peaks, which have been related to the recurrence time of classical orbits, but it does change their relative intensity, which classically should be related to the decay time or Lyapunov exponents of the orbits.[12] Apart from background ripples the first major feature that appears in all of the spectra is at $T \sim 1.5T_c$ and corresponds to a spacing of $\sim 0.67\omega_c$ ($\omega_c = 5.7$ cm^{-1} for $B = 6.1$ T). This is the first of the quasi-Landau modulations, which gives a spacing of $\sim 0.64\omega_c$ at the ionization threshold. Its first harmonic appears at $T \sim 3.0T_c$. There is also a peak at $T \sim 3.42T_c$ which corresponds to a spacing of $0.29\omega_c$. The $0.39\omega_c$ spacing does not appear with any significant intensity in any of the spectra. This spacing was present in the experimental spectrum in hydrogen,[8] but both the initial-state, H$(2p)$, and the final-state symmetries reached were different from those considered in this paper. The intensities of the quasi-Landau modulations appear not only to depend on the excitation mechanism but also to be very sensitive to the strength of the magnetic field.[7] Despite the differences between the hydrogen and lithium absorption spectra in Fig. 2 Fourier-transform spectra in Figs. 3(a) and 3(b) are almost identical. This is because the differences between the absorption spectra are only significant over small energy intervals and so the differences between the two atoms only becomes apparent if we take the transform to longer times. This is true also for strontium.

In conclusion, we have calculated the photoabsorption spectra for lithium and strontium in a magnetic field of 6.1 T in the vicinity of their ionization thresholds. We have shown that quasi-Landau modulations can be observed in such nonhydrogenic systems and that the effect of the core is to alter their relative intensity. It would be of interest to see if semiclassical theories[4,12] also give these intensity variations.

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