Perturbation theory for Rydberg states

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A perturbation theory derived for highly excited states is applied to the hydrogen atom in a magnetic field. The energy density of the states and their polarizability are calculated.

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In a well-known approach in classical mechanics, one distinguishes between fast and slow motions[1,2] in order to reduce the problem to a simpler one, i.e., to obtain an average Hamiltonian for the slow motion. In the present letter we propose an analogous procedure for highly excited states in a discrete spectrum. If variables can be separated for the unperturbed system, then simple rules can be found for determining the changes caused in the level energies by perturbations and for finding the matrix elements of physical quantities. Of particular interest in connection with the problem of the hydrogen atom is the case of classically degenerate frequencies. We will apply the theory to the problem of a hydrogen atom in a magnetic field.

The linear Zeeman effect causes a level shift $m\omega_c/2$, where m is the azimuthal quantum number, and ω_c is the cyclotron frequency. We are then left with an $(n-m)$-fold level degeneracy, which is removed by the quadratic effect. The quadratic Zeeman effect for Rydberg states was studied experimentally in Refs. 3–5, where a pseudocrossing of levels was found in a magnetic field. A theory for the quadratic Zeeman effect was derived in Refs. 6–11. The semiclassical quantization conditions were found there for the case with a quadratic Zeeman effect; the only distinguishing feature of these quantization conditions is the particular choice of variables. In the present letter we report calculations of the state density and polarizability of a Rydberg atom in a magnetic field. The Hamiltonian of the problem is (we are using atomic units)

$$H = H_0 + \frac{m\omega_c}{2} + V, \quad H_0 = \frac{p^2}{2} - \frac{1}{r}, \quad V = \frac{1}{8}\omega_c^2 \rho^2,$$

where $\omega_c = \mathscr{H}/c$ is the cyclotron frequency, and ρ is the component of the radius vector perpendicular to the magnetic field. Where perturbation theory is applicable $(V \ll H_0)$, the quantization condition can be written as follows in parabolic coordinates[8,9]:

$$\int_{k_1}^{k_2} dk \arccos\left(\frac{\epsilon + \frac{3}{2}k^2}{r_+(k)r_-(k)}\right) = k_2\phi_2 - k_1\phi_1 + 2\pi s, \quad \epsilon = \frac{8E}{\omega_c^2 n^2} - \frac{3}{2}k^2 + \frac{1}{2}m^2,$$

$$r_\pm^2 = (n \pm k)^2 - m^2, \tag{1}$$

where E is the level energy, reckoned from the Coulomb energy, $-1/2n^2$; n and m are the principal and magnetic quantum numbers; k is the "electrical" quantum number in parabolic coordinates; s is a new adiabatic invariant, which takes on integer values; the turning points $k_{1,2}$ are determined by the condition that the argument of the arc cosine is equal to ± 1; and the phases are $\phi_{1,2} = 0$ or π.

Quantization condition (1) is expressed in terms of the complete elliptic integral of the third kind, which poses severe difficulties for analysis. The expression for the state density $\rho(\epsilon)$ is far simpler. The states of a Rydberg atom in a magnetic field are symmetric with respect to inversion for $-n^2 + m^2 \leqslant \epsilon \leqslant n^2 - m^2$ and for any permissible values of $|m|/n$. If $|m|/n < 1/\sqrt{5}$, then some of the states with energies

$$-\tfrac{3}{2}(n^2 + m^2) + \sqrt{5}nm \leqslant \epsilon \leqslant -n^2 + m^2$$

are asymmetric and twofold degenerate[8,9,11]:

$$\rho(\epsilon) = \frac{\partial S}{\partial \epsilon} = \frac{8K(\lambda)}{\pi\omega_c^2 n^2 R}; \quad \lambda^2 = \frac{1}{2}\left(1 - \frac{2(n^2+m^2) + 3\epsilon}{R^2}\right), \tag{2}$$

where $K(\lambda)$ is the complete elliptic integral of the first kind, and

$$R^2 = 2\left\{\left[\frac{3}{2}(n^2 + m^2) + \epsilon\right]^2 - 5nm\right\}^{1/2}$$

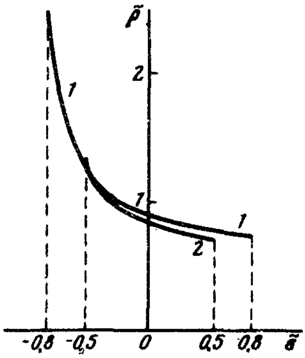

FIG. 1.

Eqauation (1) holds for symmetric states. For asymmetric states we have

$$\rho(\epsilon) = \frac{16K(1/\lambda)}{\pi\omega_c^2 n^2 \sqrt{R^2 - [2(n^2 + m^2) + 3\epsilon]}} \tag{3}$$

State density (1), (2) diverges logarithmically to infinity at $\epsilon = -n^2 + m^2(\lambda = 1)$. A stronger singularity (a root singularity) arises in the particular case $|m|/n = 1/\sqrt{5}$. Figure 1 shows the state density for $|m|/n = 1/\sqrt{2}(2)$ and $|m|/n = 1/\sqrt{5}(1)$. Figure 2 shows $\tilde{\rho}(\tilde{\epsilon})$ for the case $m = 0$. The energy variable here is $\tilde{\epsilon} = \epsilon/n^2$, and $\tilde{\rho} = \rho\omega_c^2 n^2/16$.

Let us examine the level shift in a weak electric field, $\mathscr{E} \ll \mathscr{H}^2 n^2/c^2$. As shown in Ref. 3, when a field is applied in the same direction as the magnetic field, the Stark effect is always quadratic for symmetric states or linear for asymmetric states. Calculating the average value $z = (3/2)nk$, we find

$$\mathscr{E} \langle z \rangle = \pm \frac{6}{\omega_c^2 n \rho(\epsilon)} \,. \tag{4}$$

The linear Stark shift and the state density are thus related by simple expression (4), which can be tested experimentally. In other cases there is no linear Stark effect. Let us write the expressions for the quadratic corrections to the levels, $\delta\epsilon_{\parallel}$ and $\delta\epsilon_{\perp}$, for

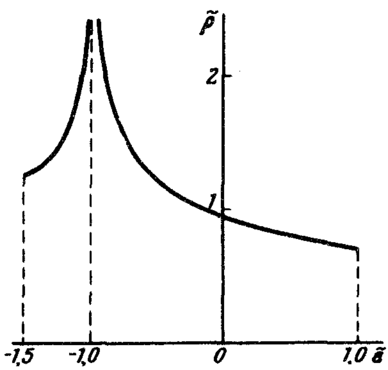

FIG. 2.

the cases in which the electric and magnetic fields are respectively parallel and perpendicular to each other. The result for the general case is extremely complicated, but for $m \ll n$ (a case of physical interest, which arises during optical excitation) the general result simplifies greatly, becoming

$$\delta\epsilon_{\parallel} = \frac{9\mathcal{E}^2}{\omega_c^2}\left(1 - \frac{4}{5}\frac{E(\lambda)}{K(\lambda)}\right), \tag{5}$$

where $E(\lambda)$ is the complete elliptic integral of the second kind. The shift is positive and inversely proportional to $\mathscr{H}^2$. A calculation of the transverse shift yields

$$\delta\epsilon_{\perp} = -\frac{9n^6\mathcal{E}^2}{32(1+4\lambda^2)}\left\{\frac{E(\lambda)}{K(\lambda)}\left[\frac{2(3+6\lambda^2+16\lambda^4)}{1+4\lambda^2} + \frac{4}{K(\lambda)\sqrt{1+4\lambda^2}}\int_0^{\lambda} j(\lambda')K(\lambda')d\lambda'\right] + 9 - 4\lambda^2\right\}, \tag{6}$$

where

$$j(\lambda) = \frac{\lambda(7-8\lambda^2-64\lambda^4)}{1+4\lambda^2}$$

The transverse shift is negative and independent of the magnetic field. The transverse polarizability is smaller by a factor $\omega_c^2 n^6 \ll 1$ than the longitudinal polarizability. We note that $\omega_c^2 n^6$ is the parameter of the classical perturbation theory. Figure 3 shows curves of (5) and (6) ($\delta\epsilon_{\parallel} = \delta\epsilon_{\parallel}\omega_c^2/9$, $\tilde{\delta\epsilon}_{\perp} = -\delta\epsilon_{\perp}\cdot 32/27n^6$).

In summary, the properties of highly excited states of the hydrogen atom in a magnetic field can be described in extreme detail. The linear Stark shift is determined by the state density. The signs of the quadratic shifts are different: The atom is a paraelectric in the longitudinal direction but a dielectric in the transverse direction.

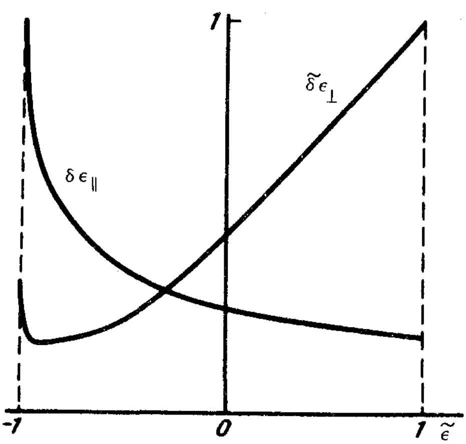

FIG. 3.

The calculation method will be described in more detail in a separate paper.

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