Asymmetric-top description of Rydberg-electron dynamics in crossed external fields

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(Received 11 September 1992)

Rydberg atoms in external electric and magnetic fields have recently emerged as atomic-scale laboratories where the quantum mechanics of classically nonintegrable systems can be studied. In this article we show, using classical canonical perturbation theory, how the intricate nonlinear dynamics of a Rydberg electron in crossed electric and magnetic fields can be described in terms of two coupled asymmetric tops.

PACS number(s): 32.60.+i, 03.65.Sq, 31.10.+z

I. INTRODUCTION

The hydrogen atom in perpendicular static electric and magnetic fields is one of the classic problems of early atomic theory. In the 1920s, an elegant treatment combining advanced methods of celestial mechanics with the old quantum theory yielded a first-order energy expression which brought out the double degeneracy in this system [1]. In refined form, this expression has been used by spectroscopists to analyze energy levels in the limit of weak fields [2,3]. Interest in this system revived following independent suggestions by Burkova *et al.* [4] and Rau [5] that the very different effects of the two fields may cooperate to stabilize the atomic electron at a considerable distance from the nucleus, thus creating unusual states [6–8]. Recently, nonlinear dynamicists have joined atomic physicists in researching the crossed-fields problem because Rydberg states in external fields are now universally recognized as microscopic laboratories in which the quantum mechanics of classically chaotic systems can be investigated [9–11].

The deceptive simplicity of the corresponding perturbed Coulomb Hamiltonian is belied by the rich nonlinear dynamics it generates. Great complexity is also evident in the recent high-resolution experiments of Holle, Wiebusch, and Welge [12] and Raithel, Fauth, and Walther [13], who studied the spectra of Rydberg electrons placed in crossed fields. Their striking success in relating peaks of their spectra to periodic orbits of the Hamiltonian has not diminished the wealth of information which still awaits analysis. Consequently there is great need for approaches that can uncover simpler structures that support this complexity.

In the present article, we demonstrate that this intricate problem can be simplified considerably by expressing it in terms of coupled asymmetric tops. This extends recent results showing that the complex electronic spectrum of the quadratic Zeeman effect in the high-principal-quantum-number limit can be systematized in terms of the state structure of the asymmetric top [14–18]. Indeed, the past few years have seen the recognition of the asymmetric top as a paradigm underlying certain localization phenomena [19], and our derivation places the crossed-fields problem among such physical scenarios recently reviewed by Rau [20].

In our derivation we use canonical perturbation theory [21]. This procedure, originally devised for problems of celestial mechanics [21], can be a highly effective tool when used in combination with the apt action-angle variables [22] of the unperturbed problem [23,24]. Recently, we have demonstrated the use of extended Lissajous variables [22] to another perturbed Coulomb problem (namely the parallel-field Stark-quadratic Zeeman effect [23,24]) and we use them here also.

This article is organized as follows. First we convert the crossed-fields Hamiltonian into a pseudo-Hamiltonian on which perturbation analysis can be performed. Our dynamical variables come from the Kustaanheimo-Stiefel (KS) transformation [25–28]. Then the two angular momenta inherent to the Coulomb problem [29–32] are expressed in terms of the KS variables using the electron angular momentum **L** and the Runge-Lenz-Laplace vector **A**. When the outcome of the canonical perturbation theory is recast in terms of these angular momenta it can be recognized as a pair of degenerate asymmetric tops coupled to each other. The paper concludes with a discussion of the results.

II. HAMILTONIAN IN FOUR DIMENSIONS

For definiteness, we assume that the electric and magnetic fields are in the x and z directions, respectively. The hydrogen-atom Hamiltonian becomes in atomic units

$$H=\frac{1}{2}p^2+\frac{B}{2}L_z+\frac{B^2}{8}(x^2+y^2)-\frac{1}{r}+Fx \ , \tag{1}$$

where B is the magnetic field in units 2.35×10^5 T, and the electric field F is in units of 5.14×10^9 V cm^{-1}.

Classical perturbation theory is performed in the Kustaanheimo-Stiefel coordinates [25–28] which allow the perturbation expansion to be readily converted into an expression containing apt action-angle variables that reflect the symmetry of the original problem. The KS transformation was originally designed to regularize the effect of the Coulomb singularity on the classical dynamics in the vicinity of the nucleus. It allows the unperturbed Hamiltonian and any perturbation to be written in terms of the canonical coordinates and the momenta of an isotropic four-dimensional harmonic oscillator.

The KS transformation starts by relating the original coordinates to a set of coordinates in a four-dimensional space using [26]

$$\mathbf{r}=\mathbf{T}\mathbf{u} \ , \tag{2}$$

where

$$\mathbf{T}=\begin{bmatrix} u_1 & -u_2 & -u_3 & u_4 \\ u_2 & u_1 & -u_4 & -u_3 \\ u_3 & u_4 & u_1 & u_2 \\ u_4 & -u_3 & u_2 & -u_1 \end{bmatrix} \tag{3}$$

and $\mathbf{u}=(u_1,u_2,u_3,u_4)$, $\mathbf{r}=(x_1,x_2,x_3)$, and $\mathbf{T}$ satisfies the orthogonality relation

$$\mathbf{T}{}^t\mathbf{T}={}^t\mathbf{T}\mathbf{T}=|\mathbf{u}|^2=r \ , \tag{4}$$

where the lowercase t stands for transpose. In Eq. (4),

$$|\mathbf{u}|^2=u_1^2+u_2^2+u_3^2+u_4^2 \ . \tag{5}$$

The two sets of coordinates are related explicitly by

$$x_3=x=2(u_1u_3+u_2u_4) \ , \tag{6}$$

$$x_2=y=2(u_1u_2-u_3u_4) \ , \tag{7}$$

$$x_1=z=u_1^2-u_2^2-u_3^2+u_4^2 \ . \tag{8}$$

The dynamical variables can be related by using the momenta $\mathbf{P}_u$ conjugate to $\mathbf{u}$,

$$\mathbf{P}_u={}^t(P_1,P_2,P_3,P_4) \tag{9}$$

for which the constraint

$$u_1P_4-u_4P_1+u_3P_2-u_2P_3=0 \tag{10}$$

holds, and the additional relation

$$\sum_{i=1}^{3} p_{x_i}dx_i=\sum_{i=1}^{4} P_i du_i \ . \tag{11}$$

Thus

$$\mathbf{p}_x=\frac{1}{2r}\mathbf{T}\mathbf{P}_i \tag{12}$$

where

$$\mathbf{P}_x={}^t(p_{x_1},p_{x_2},p_{x_3}) \ . \tag{13}$$

In view of Eq. (10), the system is subject to the constraint

$$L_z=P_\phi=m=u_4P_1-u_1P_4=u_3P_2-u_2P_3 \ , \tag{14}$$

which converts the Hamiltonian to

$$H=\frac{1}{8r}\mathbf{P}_u^2-\frac{1}{|\mathbf{u}|^2}+\frac{B}{2}L_z+\frac{B^2}{2}(u_1^2+u_4^2)(u_2^2+u_3^2) +2F(u_1u_3+u_2u_4) \ . \tag{15}$$

This, in turn, can be converted into a system of four coupled anharmonic oscillators by making the transformation to a time variable s (regularization) [26],

$$\frac{dt}{ds}=4r=4|\mathbf{u}|^2 \ . \tag{16}$$

Multiplying through by $4r$ gives

$$4=\tfrac{1}{2}(\mathbf{P}_u^2+\omega^2|\mathbf{u}|^2)+2BL_z|\mathbf{u}|^2 +2B^2|\mathbf{u}|^2(u_1^2+u_4^2)(u_2^2+u_3^2) +8F|\mathbf{u}|^2(u_1u_3+u_2u_4) \ , \tag{17}$$

where

$$\omega^2=-8E \ . \tag{18}$$

Scaling the coordinates and momenta

$$u_i\to u_i\omega^{-1/2} \ , \quad p_i\to p_u\omega^{1/2} \tag{19}$$

results in the pseudo-Hamiltonian

$$\mathcal{K}=\frac{4}{\omega}=\frac{1}{2}(\mathbf{P}_u^2+|\mathbf{u}|^2)+\frac{2}{\omega^2}BL_z|\mathbf{u}|^2 +2\frac{B^2}{\omega^4}|\mathbf{u}|^2(u_1^2+u_4^2)(u_2^2+u_3^2) +\frac{8F}{\omega^3}|\mathbf{u}|^2(u_1u_3+u_2u_4) \ . \tag{20}$$

III. APT VARIABLES AND TRANSFORMATIONS

From the classical-mechanical definitions

$$\mathbf{L}=\mathbf{r}\times\mathbf{p} \tag{21}$$

and

$$\mathbf{A}=\frac{1}{(-2mH_0)^{1/2}}\left[\mathbf{p}\times\mathbf{L}-\frac{me^2}{r}\mathbf{r}\right] \tag{22}$$

for the unperturbed Coulomb Hamiltonian

$$H_0=\frac{1}{2m}p^2-\frac{e^2}{r} \tag{23}$$

the components of $\mathbf{L}$ and $\mathbf{A}$ in four-dimensional space can be obtained using the KS transformation. They are

$$\begin{aligned} L_x&=\tfrac{1}{2}[(u_2P_1-u_1P_2)+(u_4P_3-u_3P_4)] \ , \\ L_y&=\tfrac{1}{2}[(u_1P_3-u_3P_1)+(u_4P_2-u_2P_4)] \ , \\ L_z&=u_4P_1-u_1P_4=u_3P_2-u_2P_3 \ , \end{aligned} \tag{24}$$

and

$$\begin{aligned} A_x&=-\tfrac{1}{2}[(P_1P_3+P_2P_4)+(u_1u_3+u_2u_4)] \ , \\ A_y&=-\tfrac{1}{2}[(P_1P_2-P_3P_4)-(u_1u_2-u_3u_4)] \ , \\ A_z&=\tfrac{1}{4}[(P_2^2+u_2^2+P_3^2+u_3^2)-(P_1^2+u_1^2+P_4^2+u_4^2)] \ . \end{aligned} \tag{25}$$

Now, a rotation in phase space is needed:

$$u_1=\frac{1}{\sqrt{2}}(q_1+q_4)\ ,\quad P_1=\frac{1}{\sqrt{2}}(p_1+p_4)\ ,$$
$$u_2=\frac{1}{\sqrt{2}}(q_2+q_3)\ ,\quad P_2=\frac{1}{\sqrt{2}}(p_2+p_3)\ ,$$
$$u_3=\frac{1}{\sqrt{2}}(p_2-p_3)\ ,\quad P_3=\frac{1}{\sqrt{2}}(q_3-q_2)\ ,$$
$$u_4=\frac{1}{\sqrt{2}}(p_4-p_1)\ ,\quad P_4=\frac{1}{\sqrt{2}}(q_1-q_4)\ . \tag{26}$$

Next, we seek a transformation to action-angle variables. Thereby the normal form resulting from the perturbation treatment will be converted to an expression containing the actions m and A_z and their conjugate angles ϕ_m and ϕ_{A_z}, as well as the conserved action n. The initial transformation is

$$q_i=\sqrt{2I_i}\sin\phi_i\ ,\quad p_i=\sqrt{2I_i}\cos\phi_i\ , \tag{27}$$

where $i=1,2,3,4$. Two further transformations are then performed in order to eliminate two angles. First,

$$I_1=(I_a+I_b)\ ,\quad \phi_1=\tfrac{1}{2}(\phi_a+\phi_b)\ ,$$
$$I_2=(I_a-I_b)\ ,\quad \phi_2=\tfrac{1}{2}(\phi_a-\phi_b)\ ,$$
$$I_3=(I_c+I_d)\ ,\quad \phi_3=\tfrac{1}{2}(\phi_c+\phi_d)\ ,$$
$$I_4=(I_c-I_d)\ ,\quad \phi_4=\tfrac{1}{2}(\phi_c-\phi_d)\ . \tag{28}$$

The constraint (14) requires that

$$I_a=I_c=n\ . \tag{29}$$

The final transformation of the Lissajous action-angle variables is

$$I_b=m+A_z\ ,\quad \phi_b=\tfrac{1}{2}(\phi_m+\phi_{A_z})\ ,$$
$$I_d=m-A_z\ ,\quad \phi_d=\tfrac{1}{2}(\phi_m-\phi_{A_z})\ . \tag{30}$$

The two angular momenta

$$\mathbf{J}=\tfrac{1}{2}(\mathbf{L}+\mathbf{A})\ ,\quad \mathbf{K}=\tfrac{1}{2}(\mathbf{L}-\mathbf{A}) \tag{31}$$

are the Lie-algebraic generators of SU(2)⊗SU(2) [which is locally isomorphic to the symmetry group of the unperturbed Coulomb problem, SO(4)] [18,29–32] and provide the most direct route to the asymmetric top description we are seeking. Note that the components of the vectors **J** and **K** take particularly simple forms under this succession of transformations, viz.

$$J_x=\tfrac{1}{2}[n^2-(m+A_z)^2]^{1/2}\sin(\phi_m+\phi_{A_z})\ ,$$
$$J_y=-\tfrac{1}{2}[n^2-(m+A_z)^2]^{1/2}\cos(\phi_m+\phi_{A_z})\ , \tag{32}$$
$$J_z=\tfrac{1}{2}(m+A_z)\ ,$$

and

$$K_x=-\tfrac{1}{2}[n^2-(m-A_z)^2]^{1/2}\sin(\phi_m-\phi_{A_z})\ ,$$
$$K_y=\tfrac{1}{2}[n^2-(m-A_z)^2]^{1/2}\cos(\phi_m-\phi_{A_z})\ , \tag{33}$$
$$K_z=\tfrac{1}{2}(m-A_z)\ .$$

J and **K** obey the angular momentum Poisson bracket relations. Note that

$$\mathbf{J}\cdot\mathbf{J}=\mathbf{K}\cdot\mathbf{K}=\frac{n^2}{4}\ , \tag{34}$$

which in the correspondence principle limit is slightly different from their length in quantum mechanics [29–32]:

$$\mathbf{J}\cdot\mathbf{J}=\mathbf{K}\cdot\mathbf{K}=\frac{n^2-1}{4} \tag{35}$$

since

$$\mathbf{A}\cdot\mathbf{A}=\mathbf{L}\cdot\mathbf{L}=n^2-1\ . \tag{36}$$

IV. PERTURBATION THEORY

To obtain the normal form, we use procedure due to Birkhoff [33–36], as modified by Gustavson [35] for resonant cases (as ours is). The procedure consists of finding successively better action-angle variables starting from a nonlinear Hamiltonian such as Eq. (20). It is suitable for automatic manipulation [35,36], and we have used a symbolic manipulation routine written in the MATHEMATICA language to obtain the normal form [37]. The interested reader is referred to this paper [37] where the details of the computation are explained.

One technical aspect of the problem needs to be emphasized here: Since the Coulomb Hamiltonian is experiencing two distinct perturbations, a multiple perturbation treatment is needed. In conventional quantum-mechanical treatments, the dependence of the energy on the fields changes according to the choice of the unperturbed Hamiltonian as well as the relative field strengths [38-41]. In contrast, the normal form theory employed in this paper prescribes a unique ordering of perturbations according to powers of the displacements [37].

V. RESULTS

The normal form of the pseudo-Hamiltonian, correct to second order in the angular momenta **J** and **K**, is

$$\begin{aligned}\mathcal{H}^{\rm NF}=\frac{4}{\omega}=2n&+\frac{4Bn}{\omega^2}(J_z+K_z)+\frac{24Fn}{\omega^3}(J_x-K_x)\\&+\frac{B^2}{\omega^4}[6n^3-16n(J_xK_x+J_yK_y)-12n(J_x^2+K_x^2)]\\&+\frac{FB}{\omega^5}[128(J_zK_x-J_xK_z)]\\&+\frac{F^2}{\omega^6}[960nJ_xK_x-336n(K_x^2+J_x^2)-136n^3]\ .\end{aligned} \tag{37}$$

From this expression, $2n$ can be expressed as a power series of ω^{-1}, and the energy can be obtained as a function of n, F, B, and the various angular momentum components from this series by reversion [42]. The resulting energy expression is

$$E^{(2)}=-\frac{1}{2n^2}+\frac{B}{2}(J_z+K_z)-\frac{3Fn}{2}(J_x-K_x)+\frac{B^2n^2}{16}[3n^2-4(J_z^2+K_z^2-J_zK_z)-8(J_xK_x+J_yK_y)]$$
$$+\frac{F^2n^4}{16}[-17n^2+12(J_x^2+K_x^2+J_xK_x)]+\frac{FB}{2}n^3(J_zK_x-J_xK_z)\ . \tag{38}$$

This is the desired result which shows how the crossed fields couple the angular momentum vectors **J** and **K** (remember that n is conserved). If the purely quadratic terms are examined, it becomes clear that the two asymmetric tops contained in the expression of the form

$$H^{AT}=AJ_x^2+BJ_y^2+CJ_z^2 \tag{39}$$

are degenerate and are coupled through bilinear terms containing their components.

Clearly, by using the vectors [39,40,43]

$$\mathbf{\Omega}_1=\tfrac{1}{2}(\mathbf{B}-3n\mathbf{F})\ ,\quad \mathbf{\Omega}_2=\tfrac{1}{2}(\mathbf{B}+3n\mathbf{F})\ , \tag{40}$$

both of which have magnitude

$$\Omega=\tfrac{1}{2}(B^2+9n^2F^2)^{1/2}\ , \tag{41}$$

for perpendicular fields, the resonant nature of the problem can be brought out in the first-order contribution to the energy [2,43]

$$E^{(1)}=\mathbf{\Omega}_1\cdot\mathbf{J}+\mathbf{\Omega}_2\cdot\mathbf{K}\ . \tag{42}$$

This resonance in the first-order contribution is destroyed when the two fields are not perpendicular.

For some quantum-mechanical applications [2,43], the Hamiltonian implied in Eq. (42) is transformed using the rotation operator

$$\exp[i\Omega(\mathbf{J}+\mathbf{K})\cdot\hat{\mathbf{n}}]=\exp(i\Omega A_y) \tag{43}$$

with $\hat{\mathbf{n}}=\hat{\mathbf{y}}$. This rotation simplifies the first-order energy contribution to

$$E^{(1)}=\Omega(J_{z'}+K_{z''})\ , \tag{44}$$

where $J_{z'}$ and $K_{z''}$ are the components of **J** and **K** along the directions of $\mathbf{\Omega}_1$ and $\mathbf{\Omega}_2$, respectively. However, the second-order expression is thereby complicated considerably, and has therefore been omitted.

VI. DISCUSSION AND OUTLOOK

If there is only one field present the second-order results agree with the second-order Stark and quadratic Zeeman results obtained before [24]. For instance, for no electric field, the contribution corresponding to the quadratic Zeeman effect can be written as

$$E_D^{(2)}=\frac{B^2n^2}{16}[3n^2-(m+A_z)^2-(m-A_z)^2$$
$$+2X\cos2\phi_{A_z}+(m^2-A_z^2)]\ . \tag{45}$$

In terms of

$$X\equiv\{[n^2-(m+A_z)^2][n^2-(m-A_z)^2]\}^{1/2}\ , \tag{46}$$

this expression becomes

$$E_D^{(2)}=\frac{B^2n^2}{16}[3n^2-m^2-3A_z^2+2X\cos2\phi_{A_z}]\ , \tag{47}$$

which, using the classical-mechanical version [24] of the Solov'ev-Herrick invariant Λ [44],

$$n^2\Lambda=2X\cos2\phi_{A_z}-2A_z^2-2m^2+2n^2\ , \tag{48}$$

is the semiclassical diamagnetic energy

$$E_D^{(2)}=\frac{B^2n^2}{16}(n^2+m^2+n^2\Lambda) \tag{49}$$

and agrees with previous results [24]. Similarly, for no magnetic field, and the electric field in the x direction, the second-order contribution is

$$E_S^{(2)}=\frac{F^2n^4}{16}[-17n^2+12(J_x^2+K_x^2+J_xK_x)] \tag{50}$$

$$=\frac{F^2n^4}{16}\{-17n^2+3[(L_x+A_x)^2+(L_x-A_x)^2+(L_x^2-A_x^2)]\} \tag{51}$$

$$=\frac{F^2n^4}{16}[-17n^2+9L_x^2+3A_x^2] \tag{52}$$

which, in turn, agrees with the previous parallel-field classical results [24].

The Hamiltonian in Eq. (38) is particularly suitable for dynamical and localization studies. It can be seen that the first-order part is integrable because it is separable, whereas $E^{(2)}$ has in its second-order contributions the nonlinearities and couplings typical of coupled asymmetric tops. There exists an analytical solution for the dynamics of an asymmetric top with linear perturbations in its components [45]; however, the higher-order couplings are more difficult to analyze. The chaotic behavior of a rotational system with similar bilinear couplings in two angular momenta has been studied by Feingold and Peres [46]. Furthermore, intricate graphical representations for such systems have been devised by Harter [47]. Analysis of the crossed-fields problem along these lines is currently in progress.

ACKNOWLEDGMENTS

We gratefully acknowledge the support of the National Science Foundation through its Grant No. CHE89-22420, and useful conversations with J. L. Wood.

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