Quasi-Penning Resonances of a Rydberg Electron in Crossed Electric and Magnetic Fields

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It is shown that the combination of crossed electric and magnetic fields and the Coulomb field of the atomic nucleus can lead to the localization of the Rydberg electron in the vicinity of the Stark saddle point. The localization principle is shown to be similar to the one which serves as the basis for a Penning trap. The localized electron is expected to give rise to quasibound states near and above the saddle-point ionization limit. These states are expected to cause modulations in the threshold photoionization cross sections.

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The effects of external electric and magnetic fields on highly excited atoms have recently attracted both experimental[1] and theoretical[2] interest. One of the more striking features of the effect of external fields is the occurrence of threshold oscillations in the photoionization cross section.[3] These oscillations have been attributed to localized electron states with energies near the ionization limit, whose confinement is due to the competition between atomic and external forces.[4] To the present this phenomenon has been observed in cases where only a magnetic or an electric field was applied. In this Letter we argue for the existence of a new type of localized electron state that exists in the presence of perpendicular electric and magnetic fields. The confinement of the Rydberg electron in a region about the Stark saddle point is described by analogy with the confinement of a charged particle by a Penning trap.[5,6]

Our analysis of this problem follows from the well known result of classical electrostatics that electric fields alone cannot trap a charged particle in a region of space free of other charges (Earnshaw's theorem).[7] However, a combination of electric and magnetic fields can confine a charged particle, with the magnetic field serving to stabilize motion along the directions of electrostatic instability. This is the basis for a number of macroscopic charged-particle traps.[6,8] Here we shall argue that, by use of strong but attainable magnetic fields, it may also be possible to develop a microscopic trap for a Rydberg electron. The net electric field in this case is provided by the Coulomb field of the nucleus and a uniform external electric field.

A particle trap using static fields must confine the electron about a point where the net electric force vanishes (i.e., a point of stable or unstable equilibrium). For a Rydberg atom in an electric field there is only one such point, the Stark saddle point. We therefore consider the equations of motion in coordinates centered about this point rather than about the atomic nucleus (see Fig. 1). In this coordinate system the electrostatic potential is given by

$$\Phi = e/[(x+x_0)^2+y^2+z^2]^{1/2} + E(x+x_0), \qquad (1)$$

where the applied electric field $\mathbf{E} = -E\hat{\mathbf{x}}$. A harmonic approximation of the potential in the region about the saddle point is sufficient to illustrate the key features of classical electron motion. For small x, y, and z, Eq. (1) is approximated by

$$\Phi = -V_c/e - (e/2x_0^3)(z^2+y^2-2x^2), \qquad (2)$$

where V_c is the energy of the classical ionization limit in the presence of the electric field. [In comparison, the corresponding potential for a Penning trap is proportional to $(2z^2-y^2-x^2)$.] Given Eq. (2), the addition of a magnetic field in the z direction and its accompanying Lorentz force leads to the following equation of motion for the electron:

$$d^2x/dt^2 = 2\omega_a^2 x - \omega_c\, dy/dt, \qquad (3a)$$

$$d^2y/dt^2 = -\omega_a^2 y + \omega_c\, dx/dt, \qquad (3b)$$

$$d^2z/dt^2 = -\omega_a^2 z, \qquad (3c)$$

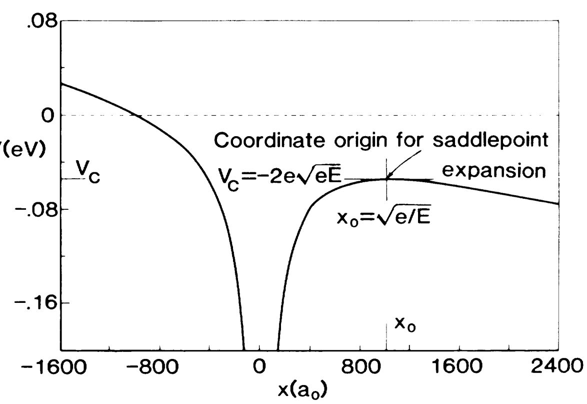

FIG. 1. Potential energy for an electron in a Coulomb field and a uniform external electric field. Here $y=z=0$, $V=-e^2/x-eEx$, $E=5$ kV/cm, and $a_0 = 5.29\times10^{-9}$ cm (Bohr radius).

where the axial frequency $\omega_a = (e^2/mx_0^3)^{1/2}$ and the magnetic cyclotron frequency $\omega_c = eB/mc$. It is evident that motion along z is harmonic. The transverse motion (i.e., in the x-y plane) can be determined by the substitution $x = a\exp(i\omega_t t)$, $y = b\exp(i\omega_t t)$. This yields

$$\omega_t^4 + (\omega_a^2 - \omega_c^2)\omega_t^2 - 2\omega_a^4 = 0, \tag{4a}$$

$$b = [i\omega_t\omega_c/(\omega_a^2 - \omega_t^2)]a. \tag{4b}$$

When $\omega_t^2 > 0$ the electron motion is periodic in an elliptical orbit. Such orbits give rise to electron states which are localized about the saddle point and whose spectrum is that of a harmonic oscillator. Divergent hyperbolic trajectories are obtained in the case when $\omega_t < 0$. Equation 4(a) leads to both types of solutions; therefore the periodic orbits are unstable with respect to small perturbations. These orbits thus assume the character of resonances rather than that of true bound states. (For the Penning trap, on the other hand, ω^2 is always positive and the motion is completely confined.)

The energy spectrum associated with the periodic orbits is that of a harmonic oscillator, namely,

$$E_{n_a,n_t} = V_c + (n_a + \tfrac{1}{2})\hbar\omega_a + (n_t + \tfrac{1}{2})\hbar\omega_t, \tag{5}$$

where n_a and n_t are respectively the quantum numbers of the axial and transverse oscillations. This describes two series of resonances spaced equally in energy, starting at the ionization energy V_c. For convenience of discussions we refer to these as quasi-Penning resonances. For the range of field strengths accessible to experiment, ω_t is comparable in magnitude to the cyclotron frequency ω_c. For instance, in fields of $E = 5$ kV/cm and $B = 150$ kG the values of $\hbar\omega_c$, $\hbar\omega_t$, and $\hbar\omega_a$ correspond respectively to 14.0, 13.2, and 6.8 cm^{-1}.

As a check on the validity of the harmonic approximation, exact classical trajectories for the electron-proton system have been computed for $E = 5$ kV/cm and $B = 150$ kG. The equations of motion were integrated numerically by use of a third-order Taylor series. It was found that for each initial position near the saddle point, an initial velocity could be found which led to a closed orbit around the saddle point. (In addition, more complicated closed orbits, such as figure 8's around the nucleus and saddle point, were found to exist.) Figure 2 shows a number of the closed orbits superimposed on the electrostatic potential-energy surface. The magnetic field here points out of the page. Agreement between our numerical calculation and our harmonic approximation [Eqs. (2)–(4)], even for the largest of the orbits shown, is good to within 5%. Thus, the anharmonic corrections to Eqs. (2)–(4) do not have a disruptive effect in this range of energies, although they do produce a slight distortion of the elliptical orbits.

We have carried out a semiclassical quantization of

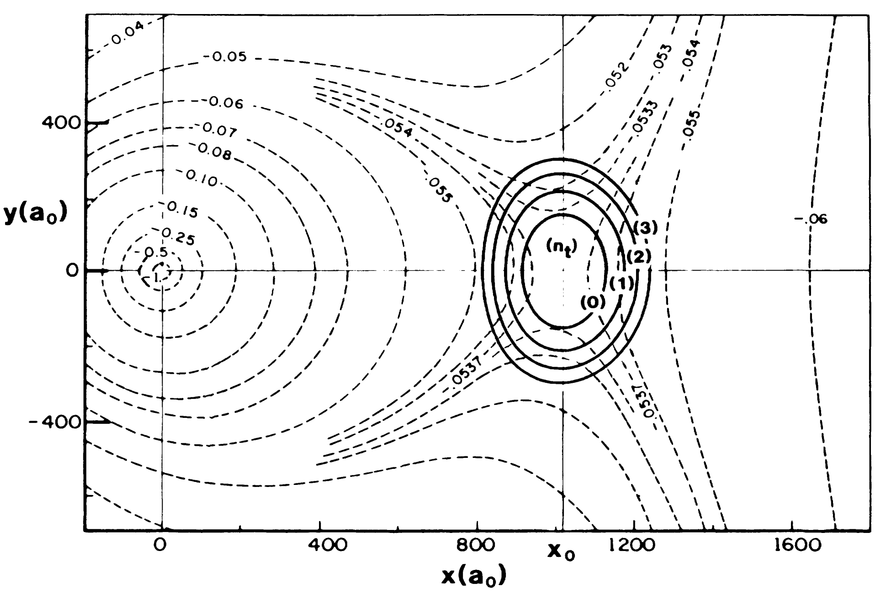

FIG. 2. Potential-energy surface and trajectories of quasi-Penning resonances. The dashed contour lines map out the potential-energy surface (in electronvolts) of the combined Coulomb and external electric field, $E = 5$ kV/cm (along $-\hat{\mathbf{x}}$). The solid lines indicate the orbits of the lowest four resonances about the saddle point for $B = 150$ kG (along $\hat{\mathbf{z}}$) labeled by the quantum number n_t. The atomic nucleus is centered at $x = y = 0$ and the saddle point occurs at $x = x_0$, $y = 0$.

the action integral for the computed orbits. In fact, the four orbits shown in Fig. 2 represent the quantized orbits corresponding to quantum numbers $n_t = 0$, 1, 2, and 3 in Eq. (5). Thus for these field strengths we anticipate the existence of several evenly spaced resonances above the saddle-point energy.

The saddle point is a critical region of space for this system, because an electron with just sufficient energy to escape from the nucleus must traverse the saddle-point region. Therefore, the presence of quasi-Penning resonances in the saddle-point region provides a mechanism for modulation of the spectrum near the ionization limit, namely, the temporary capture of a highly excited electron and its subsequent release on either side of the saddle surface. Similar processes exist in photoionization of atoms in magnetic fields,[4] in excitation and ionization of atoms by electron impact,[9] and in threshold chemical reactions.[10]

Previous treatments of the effects of crossed fields on atoms[11] have suggested that the combination of electric and magnetic forces could lead to localization of the electron in a region far from the nucleus. However, the unique importance of the ionization saddle point was not appreciated previously. These earlier treatments described this problem in terms of motion in a double-well potential, one well being due to the Coulomb potential, and another well located at a large distance from the nucleus being due to the sum of the external electric potential and the diamagnetic interaction. This picture relies upon the use of a particular gauge, and the position of the outer well turns out to depend upon the gauge used. This gauge dependence, which cannot be correct physically, is due to the neglect of a velocity-dependent term in the Hamiltonian (the paramagnetic interaction) when the problem is cast in terms of motion in a fixed potential. In fact, a full treatment of electron motion in this type of system indicates that the effects of paramagnetism are of comparable importance to those attributed to diamagnetism.[12]

The approach set forth in this Letter avoids artificial separation of diamagnetic and paramagnetic interactions by explicitly treating the full Lorentz equation of motion. This method has yielded, for the first time, a prediction of the spectrum of localized states in the semiclassical approximation. The determination of the lifetimes of these states and their associated transition moments awaits a full quantum mechanical treatment. The distinctive character of this portion of the spectrum makes it an attractive target of experimental investigation. While there have been a number of previous experimental investigations of Rydberg atoms in crossed electric and magnetic fields,[13] none yet has focused attention on the energy region near the ionization threshold. We are now in the process of conducting such an experimental study.

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