Hydrogen Atom in Crossed Magnetic and Electric Fields

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The first studies with the hydrogen atom in strong crossed magnetic and electric fields are reported. Long-living states reaching well above the Stark saddle energy are discovered. They are quantitatively explained by second-order perturbation theory, indicating regular, nonchaotic dynamics in these states. Fourier-transformed spectra and classical trajectory calculations indicate the existence of periodic orbits high in the field-ionization regime. Sensitive spectral dependence upon the angle between the fields due to symmetry breaking is observed and accounted for by second-order perturbation theory.

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The physics of the highly excited hydrogen atom in magnetic fields has recently attracted much attention and has been substantially advanced by experimental[1,2] and theoretical studies.[3,4] Here we report the first experiments with the atom in crossed magnetic and electric fields, another principally important yet incompletely solved problem.

The Hamiltonian in atomic units is

$$H=\tfrac{1}{2}\mathbf{p}^2-1/r+\tfrac{1}{2}BL_z+\tfrac{1}{8}B^2(x^2+y^2)-Fx\,, \qquad (1)$$

where the magnetic and electric fields are taken along the z and the x axis, respectively. This Hamiltonian differs from that of the hydrogen atom in a single uniform field (or two parallel fields) because L_z does not commute with the Hamiltonian. The total energy, E, and the parity with respect to the $z=0$ plane, π_z, are the only known constants of motion.

Previous spectroscopic studies have been performed with nonhydrogenic atoms and are limited to either both or one of the external fields being weak.[5-8] Quasi-Landau modulations with spacing $\frac{1}{2}\hbar\omega_c$ have been observed (though with random-motion-induced weak electric fields) and theoretically explained.[5] Very little is known about crossed-field atoms in the strong-field regime where both external forces are comparable with the Coulomb force. Aside from the general importance to atomic physics, such systems raise two questions of particular interest: One is the possibility of localizing electrons on long-range closed orbits, addressed in theoretical studies[9-11] and recent experiments.[8] The other one, concerning the dynamics, is whether the electron motion becomes classically chaotic, as in the case of the hydrogen atom in a magnetic field.[12] Electric field ionization may profoundly alter the situation.

As done previously,[1] the atoms are excited by pulsed tunable vacuum-ultraviolet (vuv) and uv laser radiation in two steps; $H(n=1)+\text{vuv}\rightarrow H(n=2)+\text{uv}\rightarrow H^*$. Employing linearly polarized vuv light (parallel to the B field), the $|2p,m=0\rangle$ sublevel of the $n=2$ manifold is selectively prepared. From there final-state spectra are taken by scanning the uv, also parallel polarized. The crossed atomic-laser-beam setup is shown in Fig. 1. The detector, consisting of a scintillator attached to a light guide and photomultiplier, was kept at a potential of +25 kV with respect to the fine-mesh metal grid at ground potential resulting in a field of 12 kV/cm. In order to appreciate the spectra observed, it is important to notice that atoms ionized promptly within the F field were not detected since protons and electrons are deflected with high drift velocity (e.g., $\sim 3\times10^6$ cm/s at 2 kV/cm) in the y direction and thus did not reach the detector. Only neutral H^* atoms carried on with the atomic beam and field ionized behind the grid constitute the observed signal. Since the atomic beam velocity is $\sim 2\times10^5$ cm/s and the distance between the grid and the excitation region is ~ 2.6 cm, only H^* atoms with lifetimes $>13\ \mu$s will be detected.

Experiments have been made at $B=6$ T and $F=2$, 3, and 4 kV/cm. Neglecting the magnetic field the corresponding classical field-ionization saddle-point energies are $E_{\text{sp}}=-274$, -335, and -387 cm^{-1}. Figure 2 shows two sections of the spectrum taken at $B=6$ T and $F=2975$ V/cm with a final-state excitation resolution of ~ 1.5 GHz determined by the uv-laser bandwidth. Essentially similar sharp-line spectra have been obtained at $F=2$ and 4 kV/cm. Two key observations valid for all three are the following: (a) The existence of long-living states at energies far beyond the saddle point, somehow

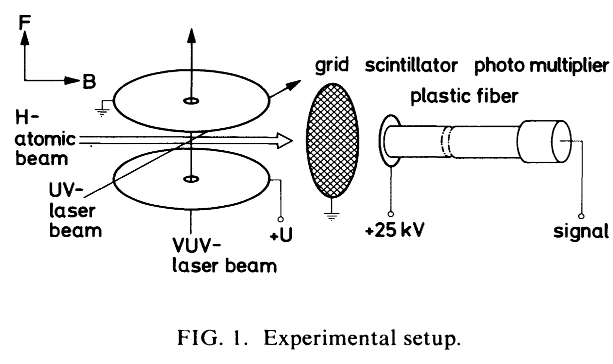

FIG. 1. Experimental setup.

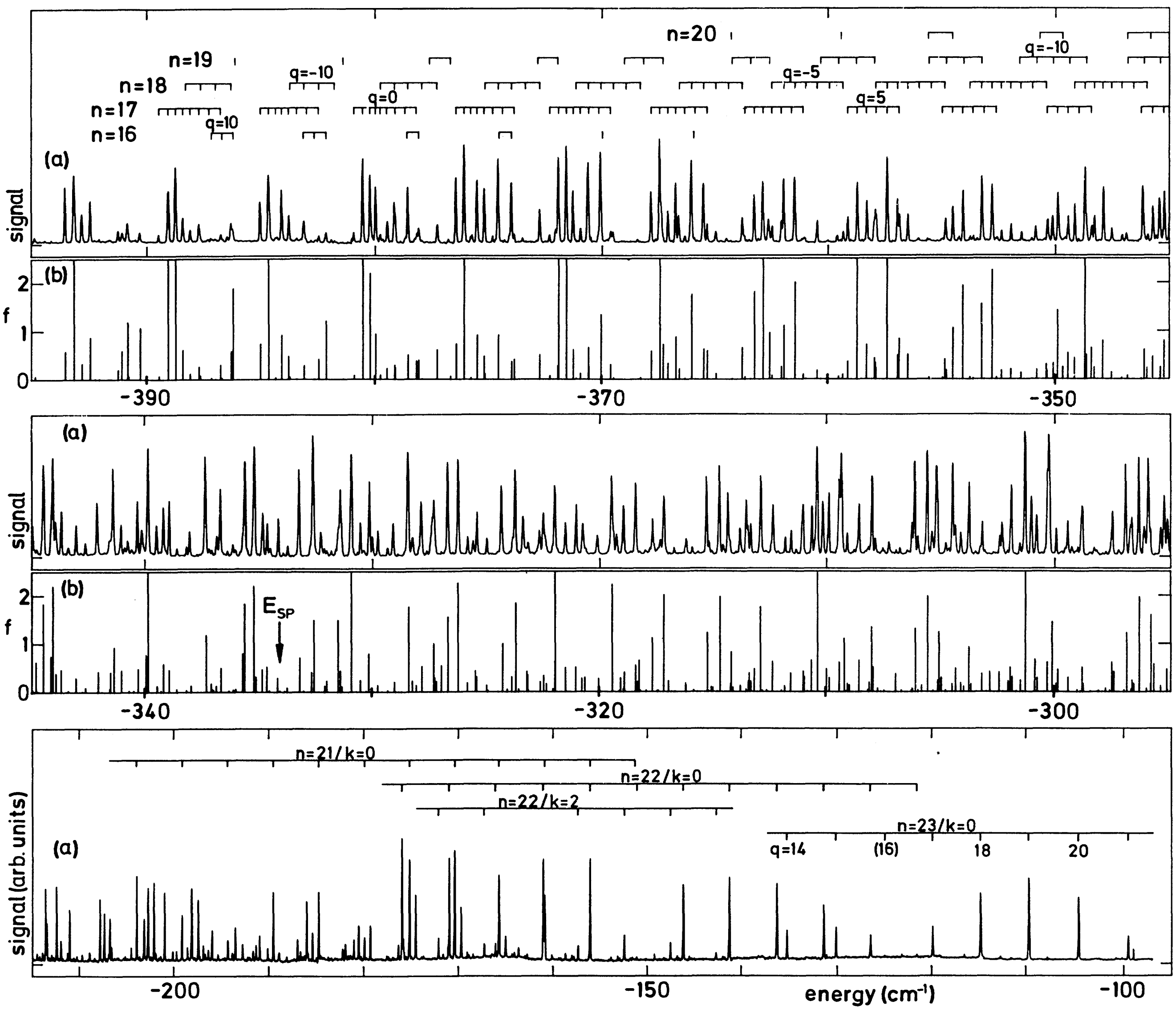

FIG. 2. Sections of a crossed-field spectra at $B=6$ T and $F=2975$ V/cm. Stark saddle-point energy $E_{sp}=-339.9$ cm^{-1}. (a) Experimental spectra; spectral resolution 0.05 cm^{-1}. (b) Theoretical spectra, calculated in second-order perturbation theory. Oscillator strength f in 10^{-5} atomic units.

reminiscent of long-living states of the H atom in electric fields;[13] (b) the transition to the ionization regime without any sign of the saddle point nor a noticeable signature or effect of a long-range potential well. Such a potential minimum has been theoretically predicted.[11] Though in principle not allowed, if one were to set, as in previous studies,[8,10,11] $L_z=0$ in Eq. (1) the resulting potential along the x axis would be $V(x)=-1/|x|+\frac{1}{8}B^2x^2-Fx$. A potential well would occur in the $F=4$ kV/cm field with a minimum at $x_0=4600a_0$ (a_0=Bohr radius). The potential with $F=2$ kV/cm would have no well, while the one at 3 kV/cm would just have a practically horizontal turning point at $x_0=3000\times a_0$.

The spectra can be well accounted for by second-order perturbation theory (SOPT). In first order, level shifts are given by[14]

$$E^{(1)}_{n,q}=\tfrac{1}{2}q(B^2+9n^2F^2)^{1/2}, \qquad (2)$$

with n the principal quantum number of the unperturbed hydrogen atom, and q the "Pauli quantum number," which runs from $-(n-1)$ to $+(n-1)$. Each level remains $(n-|q|)$-fold degenerate. This degeneracy is lifted in the SOPT developed by Braun and Solov'ev.[15] The levels belonging to fixed n and q are counted by a quantum number $k=0,1,\ldots,n-|q|-1$ starting at the highest energy value. The calculated SOPT eigenvalues, E_{nqk}, agree quite well with the experimental line positions (Fig. 2) with deviations getting larger with increasing principal quantum number n. Oscillator strengths have also been calculated in SOPT and agree qualitatively with observed intensities (Fig. 2), except for larger discrepancies in strong lines possibly due to experimental saturation. The fact that the long-living states far in the continuum region are unambiguously identified by SOPT quantum numbers has an important implication: It suggests regular dynamics of the atom in these states.

To investigate the existence of long-range closed orbits in crossed-field atoms[9-11] we have Fourier transformed the energy spectra. The results are time-domain spectra with distinct resonance structures, such as those in Fig. 3 derived from the 3-kV/cm energy spectrum. As discussed previously,[2] resonances at time T_i correlate, according to $\Delta E_i = 2\pi\hbar/T_i$, with a modulation (with periodic spacing ΔE_i) in the energy spectrum and, on the other hand, with a closed orbit (type i) with period T_i of the Rydberg electron. In their general appearance the time-domain spectra consist of groups of resonances with obvious periodic order shifting systematically to shorter times with increasing energy. In detail the spectra show complex fine structure depending strongly on the energy.

In view of the correlation between T_i resonances and closed orbits[3,4] we have performed extensive classical trajectory calculations with the proton as origin. Indeed many closed orbits can be found. However, unambiguous identification with observed T_i resonances is at present not possible.

The conservation of π_z depends on the angle θ between the two fields. Breaking the π_z symmetry allows excitation of all lines to even- as well as odd-parity final states (k even or odd), while only even ones are allowed at $\theta = 90°$. Figure 4(a) shows experimental spectra obtained at $B = 6$ T and $F = 2$ kV/cm. A deviation of $\sim 0.1°$ from 90° changes the spectral pattern within the precision of these experiments. The experimental results agree well with theoretical spectra from SOPT calculations, as shown in Fig. 4(b).

In summary, we have succeeded in investigating the spectroscopy of the highly excited hydrogen atom for the first time in perpendicular (90°) magnetic and electric fields in the strong-field regime at energies into the field-ionization region. Sharp-line spectra are observed showing the existence of long-living final states high in the field-ionization continuum. There is no specific signature in these spectra indicating the field-ionization saddle point or the supposed long-range potential well. The spectra are theoretically well accounted for by a second-order perturbation treatment, indicating nonchaotic regular dynamics in the long-living states. Fourier-transform time-domain spectra show the existence of long-range periodic orbits, also obtained by classical trajectory calculations. First angular-dependence experiments at angles off 90° show the parity-symmetry-

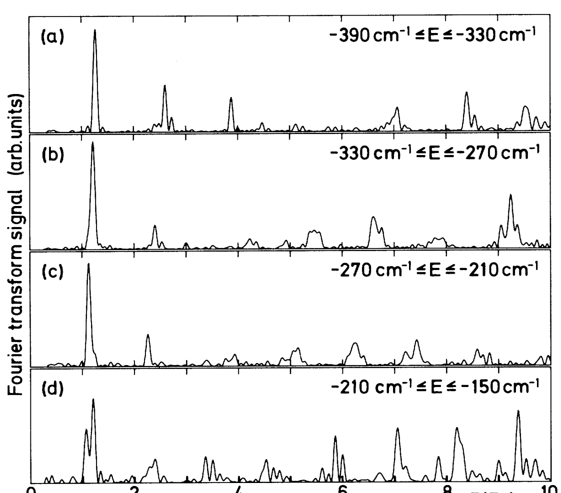

FIG. 3. Fourier-transformed spectra of experimental spectrum at $B = 6$ T, $F = 2975$ V/cm, in sections of 60 cm^{-1} width. Plotted is the absolute value squared as a function of time in units of cyclotron period $T_c = 6.0 \times 10^{-12}$ s.

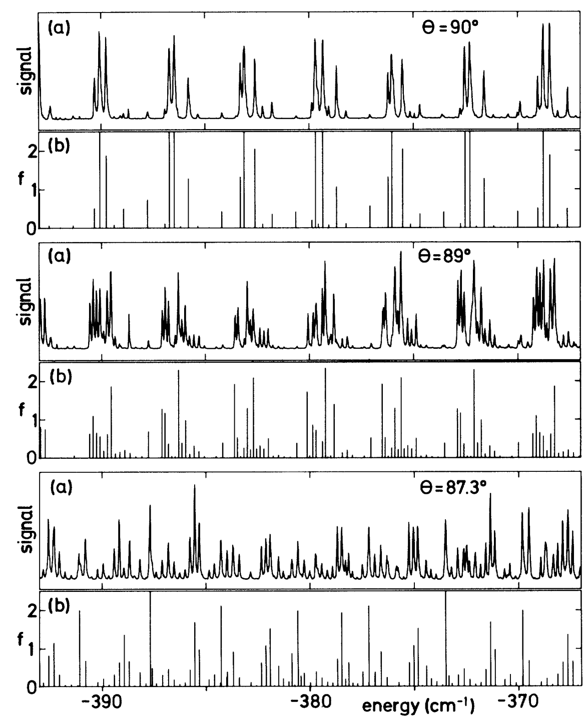

FIG. 4. Dependence of spectra on angle θ between magnetic and electric field. Field strengths: $B = 6$ T, $F = 2000$ V/cm. (a) Experimental spectra at $\theta = 90°$, 89.0°, and 87.3°. (b) Theoretical spectra in second-order perturbation theory. Oscillator strength f in 10^{-5} atomic units.

breaking effect on the spectra.

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[1]A. Holle, J. Main, G. Wiebusch, H. Rottke, and K. H. Welge, Phys. Rev. Lett. **61**, 161 (1988), and references therein.

[2]J. Main, G. Wiebusch, A. Holle, and K. H. Welge, Phys. Rev. Lett. **57**, 2789 (1986).

[3]D. Wintgen and H. Friedrich, Phys. Rev. A **36**, 131 (1987), and references therein.

[4]J. B. Delos and M. L. Du, IEEE J. Quantum Electron. **24**, 1445 (1988).

[5]K. T. Lu, F. S. Tomkins, H. M. Crosswhite, and H. Crosswhite, Phys. Rev. Lett. **41**, 1034 (1978); H. Crosswhite, U. Fano, K. T. Lu, and A. R. P. Rau, Phys. Rev. Lett. **42**, 963 (1979).

[6]E. Korevaar and M. G. Littman, J. Phys. B **16**, L437 (1983).

[7]F. Penent, D. Delande, F. Biraben, and J. C. Gay, Opt. Commun. **49**, 184 (1984); F. Penent, D. Delande, and J. C. Gay, Phys. Rev. A **37**, 4707 (1988).

[8]M. Fauth, H. Walther, and E. Werner, Z. Phys. D **7**, 293 (1987).

[9]C. W. Clark, E. Korevaar, and M. G. Littman, Phys. Rev. Lett. **54**, 320 (1985); C. Nessmann and W. P. Reinhardt, Phys. Rev. A **35**, 3269 (1987).

[10]J. C. Gay, L. R. Pendrill, and B. Cagnac, Phys. Lett. **72A**, 315 (1979).

[11]S. K. Bhattacharya and A. R. P. Rau, Phys. Rev. A **26**, 2315 (1982).

[12]A. Harada and H. Hasegawa, J. Phys. A **16**, L259 (1983); G. Wunner, U. Woelk, I. Zech, G. Zeller, T. Ertl, F. Geyer, W. Schweizer, and H. Ruder, Phys. Rev. Lett. **57**, 3261 (1986).

[13]H. Rottke and K. H. Welge, Phys. Rev. A **33**, 301 (1986).

[14]M. Born, *Vorlesungen über Atommechanik* (Springer-Verlag, Berlin, 1925); W. Pauli, Z. Phys. **36**, 335 (1926); Y. Demkov, B. S. Monozon, and V. Ostrovskii, Zh. Eksp. Teor. Fiz. **57**, 1431 (1969) [Sov. Phys. JETP **30**, 775 (1970)].

[15]E. A. Solov'ev, Zh. Eksp. Teor. Fiz. **85**, 109 (1983) [Sov. Phys. JETP **58**, 63 (1983)]; P. A. Braun and E. A. Solov'ev, Zh. Eksp. Teor. Fiz. **86**, 68 (1984) [Sov. Phys. JETP **59**, 38 (1984)].