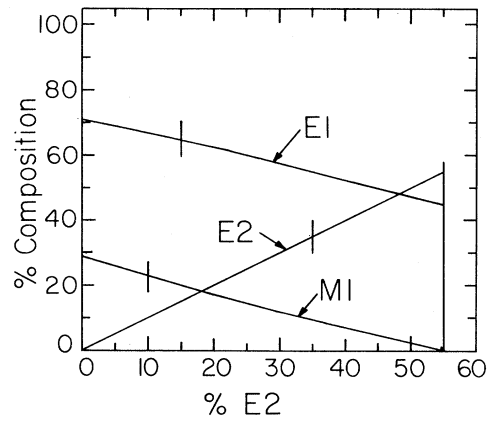

FIG. 3. Multipole composition of the statistical region (1.7–2.5 MeV) consistent with the measured conversion coefficients, shown as a function of the percent $E2$ contribution. Typical error bars are shown to indicate the experimental uncertainties.

for the high-energy part of the yrast spectrum.[6]

Our findings for the multipolarity are consistent with the angular-distribution data from Refs. 2, 3, 6, and 7 that employed heavy-ion–induced reactions, but there is an apparent disagreement with the (α, xn) work of Feenstra *et al.*[5] In the $(\alpha, 4n)$ reaction at 47 MeV the maximum angular momentum J_{max} that can be reached is ~24. In contrast, the ^{20}Ne reactions of 130 MeV give $J_{max} \approx 58$.[9] In view of this difference in angular momenta it is conceivable that the statistical cascades involved in the continuum reached by the low spins in the $(\alpha, 4n)$ reaction are different and proceed essentially by $E1$ transitions.

In summary, we have established that for the ^{20}Ne + ^{150}Nd reactions investigated, the $E1$ fraction for the statistical cascades lies in the range (45–71)%, that the middle of the yrast region is dominated by $E2$ transitions, and that the lower-energy part of the yrast region is increasingly dominated by $M1$ radiation.

The help of R. Woodward in collection and reduction of the data is appreciated. This research was supported in part by the U. S. Department of Energy. Oak Ridge National Laboratory is operated for the Department of Energy by Union Carbide Corporation.

[1]J. O. Newton *et al.*, Phys. Rev. Lett. 34, 99 (1974).
[2]M. V. Banaschik *et al.*, Phys. Rev. Lett. 34, 892 (1975).
[3]W. Trautmann *et al.*, Phys. Rev. Lett. 35, 1694 (1975).
[4]J. O. Newton *et al.*, Phys. Rev. Lett. 38, 810 (1977).
[5]F. J. Feenstra *et al.*, Phys. Lett. 69B, 403 (1977).
[6]J. O. Newton, S. H. Sie, and G. D. Dracoulis, Phys. Rev. Lett. 40, 625 (1978).
[7]R. S. Simon *et al.*, Phys. Rev. Lett. 36, 359 (1976).
[8]J. R. Grover and J. Gilat, Phys. Rev. 157, 814 (1967).
[9]D. G. Sarantites *et al.*, Phys. Rev. C 14, 2138 (1976), and 17, 601 (1978).
[10]L. Westerberg *et al.*, Nucl. Instrum. Methods 145, 295 (1977), and to be published.
[11]L. Westerberg *et al.*, Nucl. Instrum. Methods 128, 61 (1975).

Resonant States in the Nonperturbative Regime: The Hydrogen Atom in an Intense Electric Field

Peter M. Koch
Gibbs Laboratory, Yale University, New Haven, Connecticut 06520
(Received 1 May 1978)

I present the first results of a precise measurement of the intense-field Stark effect in highly excited states of atomic hydrogen, a prototype system for experimental and theoretical studies of resonant states in the nonperturbative regime. I show that the quantity n^4F (in atomic units), known to be important for field ionization, also governs the divergence properties of the perturbation series for the energy shift.

The interaction of a nonrelativistic hydrogen atom with a static electric field $\vec{F} = F\hat{z}$ is a historic problem in atomic physics. As F increases, the degenerate energy levels of each excited state fan out until, finally, the rate of decay by ionization exceeds that of spontaneous emission, and levels fade away. Each level is conveniently labeled by its parabolic quantum numbers $\{q\}$

© 1978 The American Physical Society

$=\{n, n_1, n_2, |m|\}$ which come from a zero-field separation of Schrödinger's equation in parabolic coordinates.[1]

In this Letter it is shown how the Stark effect in resolved states of H ($n \sim 25$) may be used to investigate the properties of resonances in the nonperturbative regime. The data show how, and it is explained why, Raleigh-Schrödinger perturbation theory (RSPT) can poorly approximate the energy shift for states with $n_1 - n_2 \gg 0$ but still give quite accurate results when $n_1 - n_2 \ll 0$.

Titchmarsh[2,3] has shown that the negative-energy poles of the Green's function for Schrödinger's equation with the Stark Hamiltonian, $\mathcal{H} = -\frac{1}{2}\Delta - r^{-1} + Fz$, in atomic units,[1] move off the real axis when $F > 0$. Even though there are no true bound states, the "spectral concentration" of each pole at $E_{\{q\}}(F) = -E_R(F) - i\Gamma(F)/2$ localizes a resonance centered at energy $-E_R$ that decays by ionization at a rate Γ. To describe the Stark effect, one must specify the F dependence of the complex trajectory $E_{\{q\}}(F)$ of each pole. In addition, relativistic or other effects may cause some poles on different trajectories to interact strongly when they approach each other at "level crossings."

Since the Stark resonances belong to the general class of nonstationary states found throughout quantum physics, and since their properties may be adjusted experimentally by variation of F and $\{q\}$, this problem has broad physical importance. The properties of resonances are usually calculated perturbatively.[3] In many cases it is suspected [Born series for rearrangement collisions,[4] quantum electrodynamic expansions in (α/π),[5] etc.] or known that RSPT does not converge.

Specifically, for the Stark effect RSPT yields a nonconvergent, asymptotic expansion for the real part E_R of the energy,[2,3] although nonspecialists are generally not aware of this fact. Silverstone[6] has recently used machine-computational methods to push it to at least twenty-fifth (!) order in F and has exhibited its asymptotic nature. To calculate $\Gamma(F)$, a number of authors have used the Jeffreys-Wentzel-Kramers-Brillouin and related approximations.[7] Other workers have used a variety of nonperturbative, numerical, or classical methods to obtain values for $E_R(F)$ and $\Gamma(F)$,[8] but usually only for the ground state.[9] Solid-state physicists have investigated the Stark effect for Wannier excitions, hydrogenlike systems.[10] That most of the nonperturbative methods require a high-precision, numerical integration to be made for each $\{q\}$ and F is a distinct disadvantage.

My experimental results on resolved upper and lower (always $n = 10$) Stark states of H, with energies $E_{\{U\}}(F_p)$ and $E_{\{L\}}(F_p)$, respectively, were obtained using the method of fast-beam laser spectroscopy[11-15] in an apparatus represented by Fig. 1. A hydrogen beam with a measured kinetic energy $E_b = 7474(20)$ eV included excited states weighted by n^{-3} that were produced by $H^+ + Xe$ electron-transfer collisions. A free-running, single-mode, GTE model 950 $^{12}C^{16}O_2$ laser produced a 5–25-W/cm^2 beam of photons (with energies k_l and k_R in the laboratory and atomic rest frames, respectively) which was mechanically chopped at frequency ω_1 and merged (crossing angle $\theta = 0$) with the fast atomic beam. For a given k_R, the transverse field F_p was used to tune $\{L\}$ and $\{U\}$ into resonance with the laser photons. If F_p, $E_{\{L\}}(F_p)$, and k_R were known

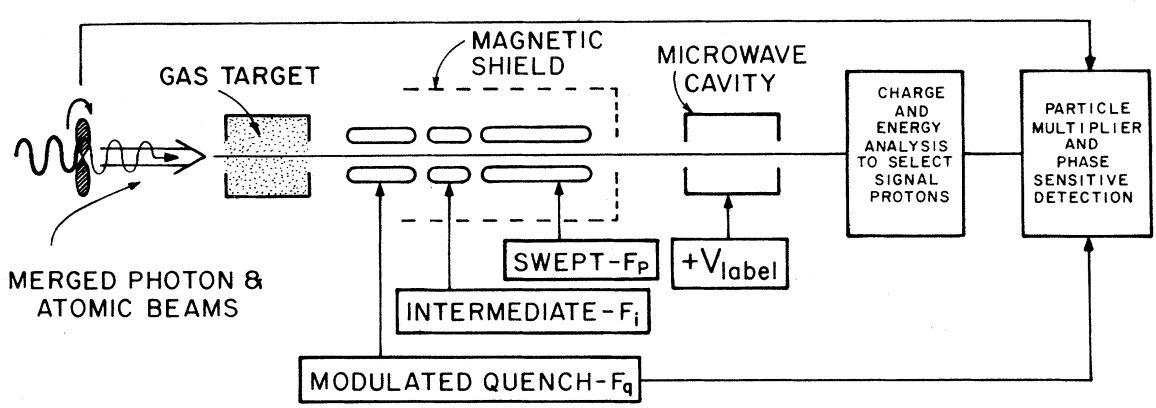

FIG. 1. Important features of the apparatus. The photon beam was linearly polarized parallel to $\vec{F}_p$ and drove $\Delta m = 0$ transitions.

absolutely, then the experiment was a measurement of $E_{\{U\}}(F_p) = k_R + E_{\{L\}}(F_p)$. Voltages applied to produce F_p were measured to <0.03%. k_l was not measured directly but determined absolutely to about 1 ppm from published values.[16,17] k_R was calculated to 5-ppm accuracy. Ignoring fine structure, which contributes <5 ppm to the energy, I used the measured value of F_p and RSPT to calculate $E_{\{L\}}(F_p)$ absolutely to ~10 ppm. This will be justified below. The experimental signal was produced by multiphoton ionization[14,18] of the laser-excited atoms, which retained their principal-quantum-number identities after leaving F_p, in a voltage-labeled TM_{010} microwave cavity[11,14,18] ($\nu = 9.91$ GHz), followed by phase-sensitive detection of the resultant energy-labeled proton current at frequency ω_1.

I have not yet worked out a detailed theory of the typical line shape shown in Fig. 2, but the asymmetry is consistent with the measured nonuniformity in the nominal 0.95-cm separation of the F_p electrodes, which were about 7 cm high and 10 cm long. For the present results the measured peak is taken to be the center of the line and the 0.1% inhomogeneity of F_p is used as an estimate of its error. The scatter of data from experimental runs on different days was typically a factor of 5 smaller. Calculated Zeeman shifts produced by measured stray (<3 mG) or motional (<30 mG) magnetic fields were less than 10^{-6} of the typical, measured Stark shifts. That we observed no shift of any resonance line when the polarity of F_p was reversed is evidence for the absence of magnetic-field and contact-potential-difference problems.

It is satisfying, but ironic, that field ionization, one of the effects being studied, provided a solution to the most challenging experimental problem —positive assignment of quantum numbers to the lines. Exposure to $F_q \sim 50$ kV/cm in the structure labeled "modulated quench-F_q" in Fig. 1 ionized atoms in only a few of the "easily ionized" $n_1 \ll n_2$ states of $n = 10$ (as well as all $n \gtrsim 11$ atoms).[7,8] Square-wave modulation of this field by ~0.5 kV/cm around F_q at frequency ω_2 and phase-sensitive detection at the beat frequency $(\omega_1 - \omega_2)$ produced a photoabsorption spectrum of only the few $n = 10$ levels whose populations were modulated.

A constant field $F_i \sim 10$ V/cm, with the same polarity as F_q and F_p, was used to avoid a region of zero field between them. Nevertheless, I observed and exploited an interesting phenomenon that I believe is caused by nonadiabatic effects in the fringe fields.[19] For example, when I set F_q near 48 kV/cm to "define" preferentially the states (10, 0, 9, 0) and (10, 0, 8, 1), I also observed spectral lines originating with the states with equal and opposite electric dipole moments, (10, 9, 0, 0) and (10, 8, 0, 1). The relative phase of the interchanged ($n_1 \gg n_2$) signals at the output of the $(\omega_1 - \omega_2)$ lockin detector, however, was always *opposite* to that of the ($n_1 \ll n_2$) signals. This provided a useful experimental signature. I verified in independent experiments that the $n_1 \gg n_2$ states were not ionized by the $F_q \sim 48$ kV/cm.[19]

In Figs. 3 and 4 experiment and theory are com-

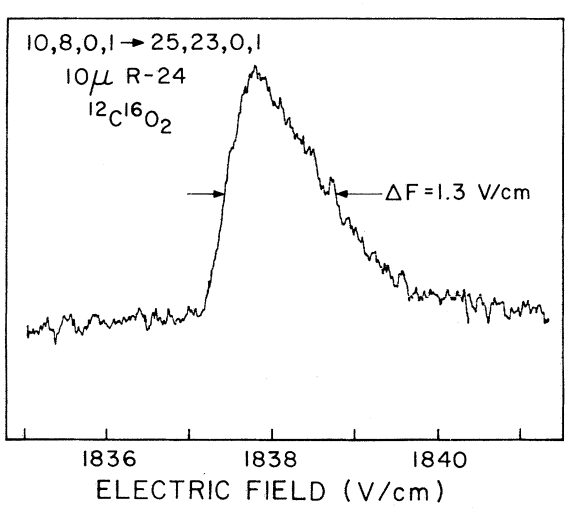

FIG. 2. A chart recording of a typical line shape.

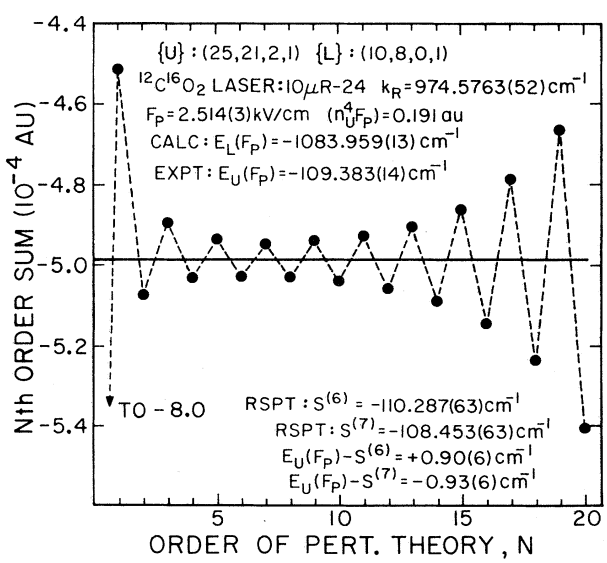

FIG. 3. Solid line: measured energy for H (25, 21, 2, 1). Closed circles: RSPT summed to order N. $\Gamma_U < 10^5$ sec^{-1}. $[2/2]_U = -108.911(60)$ cm^{-1}.

pared for E_R for the states (25, 21, 2, 1) and (30, 0, 29, 0), respectively, in the intense-field regime. The insets tabulate the measured results; after conversion to atomic units for an atom with infinite nuclear mass, the graphs display them along with RSPT summed to various orders N,

$$S^{(N)} = \sum_{i=0}^{N} E^{(i)}.$$

Propagation of the error in F_p yields the errors shown for the RSPT values. In Fig. 3, $S^{(N)}$ oscillates about the experimental result but finally diverges from it. The minimum discrepancy $D_{\min}$ between experiment and RSPT at $N=6$ is more than 10 times the estimated error. Notice that $|D_{\min}| < |E^{(7)}|$, the smallest term in the series, which is the expected behavior for oscillating, asymptotic series.[3] Moreover, for this one case at least, a simple average of $S^{(6)}$ and $S^{(7)}$ is consistent with the measured result. This may not be typical behavior. Notice that the measured energy of (25, 21, 2, 1) lies 197 cm^{-1} above the top of the barrier, $E_b = -2F_p^{1/2}$ a.u.[1,8]

In Fig. 4 the monotonic RSPT series appears to converge to the experimental result, but this is actually not the case. The smallest term is $|E^{(24)}|$; for $N>24$, $S^{(N)}$ will eventually drop below and diverge from the experimental result. The truncated sum $S^{(24)}$ agrees with the measured result within experimental error. Notice that the measured energy of (30, 0, 29, 0) lies only 3.3 cm^{-1} below the top of the barrier, $E_b = -2F_p^{1/2}$ a.u.[1,8]

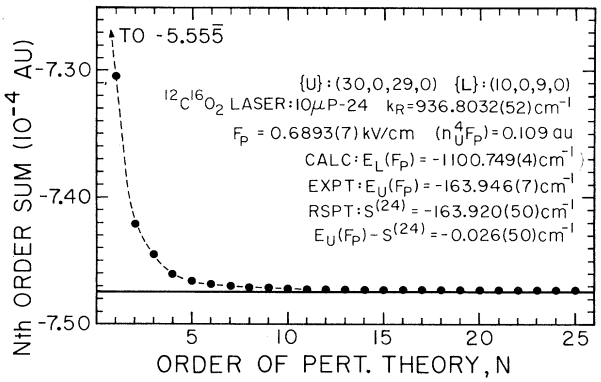

FIG. 4. Solid line: measured energy for H (30, 0, 29, 0). Closed circles: RSPT summed to order N. $\Gamma_U < 10^6$ sec^{-1}. $[2/2]_U = -163.826(47)$ cm^{-1}.

To explain the very different behavior of RSPT for the cases $n_1 - n_2 \ll 0$ and $n_1 - n_2 \gg 0$, let us examine it for H (25, 21, 2, 1) (+ signs) and H (25, 2, 21, 1) (− signs). *The series for* $2n^2E_R$ is, approximately,

$$2n^2E_R = -1 \pm 2.28(n^4F) - 1.91(n^4F)^2 \pm 3.21(n^4F)^3 - 13.0(n^4F)^4 \pm 47.9(n^4F)^5 - 236(n^4F)^6 \pm 1130(n^4F)^7 - \ldots.$$

Theory[7,8] and experiment[1,19] have shown that the ionization width $\Gamma(F)$ remains small for $n_1 - n_2 \gg 0$ resonances when $n^4F \lesssim 0.3$. Since the series shown above diverges at low order when its expansion variable $n^4F \gtrsim 0.2$, there is a range of F for which the $n_1 - n_2 \gg 0$ resonances are well localized at energies $-E_R$ whose values are poorly approximated by the oscillatory RSPT series. Figure 3 shows a case in this range. Resonant states with $n_1 - n_2 \ll 0$, however, retain small widths only when $n^4F \lesssim 0.13$. For this significantly lower range, the monotonic RSPT series does not diverge until high order and more closely approximates E_R. This behavior is shown in Fig. 4. Since $n_L{}^4F_p < 0.005$ in the present experiment, RSPT furnished accurate E_R values for all $n_L = 10$ lower states.

Since the values of n^4F above which Γ increases exponentially are similar to bounds on the domain of utility of RSPT for calculation of E_R, this parameter has a common importance for the theoretical calculation of both quantities. This is natural since E_R and Γ are the two sides of one coin.

We have also calculated the $M=1, 2$ diagonal Padé approximations[3,20] $[M/M]$ to RSPT for the $\{U\}$ resonances of Figs. 3 and 4. The $[2/2]$ value given in each figure caption is much closer to the respective measured result than is RSPT truncated at order $2M=4$.

In future experiments I shall be able to investigate the important properties of the Stark resonances at the following levels of precisions: $\Delta F_p/F_p \sim 10$ ppm; $\Delta E_R/E_R \sim 1$ ppm; $\Delta\Gamma/\Gamma \sim 1\%$; ratio r of oscillator strengths for laser-induced transitions between Stark sublevels of different n manifolds, $\Delta r/r \sim$ few percent; and the resonance line shape in the large-F regime where the Breit-Wigner parametrization of the resonance phase-shift may no longer be correct.[9] I shall report elsewhere the details of the experimental method, the investigation of level-crossing effects in the field-ionization process, and the divergence

properties of the Padé approximants.[19]

I acknowledge important conversations with R. Damburg, G. Hatton, H. Silverstone, and Yale University colleagues. R. Damburg, G. Hatton, H. Silverstone, J. Leopold, D. Bailey, and J. Hiskes kindly performed some calculations. E. Rock and H. Venkataramania assisted in the early stages of the experiment. This research was supported in part by National Science Foundation Contract No. PHY 76-84469 and by Yale University.

[1]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic, New York, 1955); E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge Univ. Press, Cambridge, 1963), Chap. 17.

[2]E. C. Titchmarsh, *Eigenfunction Expansions Associated with Second-Order Differential Equations Part II* (Oxford Univ. Press, Oxford, 1958).

[3]J. Killingbeck, Rep. Prog. Phys. 40, 963 (1977).

[4]B. H. Bransden, *Atomic Collision Theory* (Benjamin, New York, 1970), p. 166.

[5]F. J. Dyson, Phys. Rev. 85, 631 (1952).

[6]H. J. Silverstone, Phys. Rev. A (to be published), and private communication.

[7]D. S. Bailey, J. R. Hiskes, and A. C. Riviere, Nucl. Fus. 5, 41 (1965). See also the following: Quantal: R. J. Damburg and V. V. Kolosov, J. Phys. B 9, 3149 (1976); G. J. Hatton, Phys. Rev. A 16, 1347 (1977). Classical: D. Banks and J. G. Leopold, J. Phys. B 11, L5, 37 (1978), and to be published, and private communication.

[8]Damburg and Kalosov, Ref. 7; Hatton, Ref. 7; Banks and Leopold, Ref. 7.

[9]M. Hehenberger, H. V. McIntosh, and E. Brandas, Phys. Rev. A 10, 1494 (1974); C. Cerjan, W. P. Reinhardt, and J. E. Avron, J. Phys. B 11, L201 (1978), and references therein.

[10]D. F. Blossey, Phys. Rev. B 2, 3976 (1970).

[11]P. M. Koch, L. D. Gardner, and J. E. Bayfield, in *Beam-Foil Spectroscopy*, edited by D. J. Pegg and I. A. Sellin (Plenum, New York, 1976), Vol. 2.

[12]J. E. Bayfield, Rev. Sci. Instrum. 47, 1450 (1976).

[13]P. M. Koch, Opt. Commun. 20, 115 (1977).

[14]J. E. Bayfield, L. D. Gardner, and P. M. Koch, Phys. Rev. Lett. 39, 76 (1977).

[15]P. M. Koch, Bull. Am. Phys. Soc. 22, 1318, 1319 (1977), and 23, 563 (1978).

[16]T. Y. Chang. Opt. Commun. 2, 77 (1970).

[17]H. W. Mocker, Appl. Phys. Lett. 12, 20 (1968).

[18]J. E. Bayfield and P. M. Koch, Phys. Rev. Lett. 33, 258 (1974).

[19]P. M. Koch, to be published.

[20]G. A. Baker, Jr., *Essentials of Padé Approximants* (Academic, New York, 1975).

Field-Ionization Processes in Excited Atoms

Michael G. Littman, Michael M. Kash, and Daniel Kleppner
Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 10 April 1978)

We identify two different processes by which free atoms are ionized in a static electric field. The first process is by tunneling and is described accurately by theory developed for hydrogen. The second process, important for all atoms except hydrogen, is similar to autoionization and arises from the effect of level mixing. Data are presented on field ionization in lithium which display behavior due to each of these ionization mechanisms.

The problem of how an atom is ionized in an applied electric field has attracted wide theoretical[1,2] and experimental[3] interest. Considering the level of activity, it is remarkable that there is such poor agreement between rigorous theories, developed for hydrogen, and experimental observations on highly excited atoms in hydrogenic states. It is also remarkable that a simple "back-of-the-envelope" theory successfully predicts the field values at which ionization becomes significant. Our aims are to reconcile the conflicting theoretical pictures of field ionization, and to provide a useful insight into the underlying processes.

To put the problem into perspective, let us contrast two views of field ionization. In the first view, ionization occurs by tunneling of the electron through the potential barrier along the axis

© 1978 The American Physical Society