Dynamic Tunneling Ionization of Excited Hydrogen Atoms: A Precise Experiment versus Theories

B. E. Sauer, S. Yoakum, L. Moorman,[a] and P. M. Koch
Physics Department, State University of New York at Stony Brook, Stony Brook, New York 11794-3800

D. Richards and P. A. Dando
Mathematics Faculty, Open University, Milton Keynes MK7 6AA, England
(Received 25 June 1991)

New data for $n_0=24,\ldots,32$ H atoms ionized by a linearly polarized, 9.908-GHz electric field are compared with calculations. Being more precise than laser multiphoton ionization experiments with tightly bound atoms, our experiments distinguish between tunneling *through* and classical escape *over* a slowly oscillating barrier and between one- and many-state dynamical processes. Formulas used to interpret low-frequency laser multiphoton ionization data poorly describe our results. Our data delineate ranges of validity of other partly successful models and are best reproduced by a new 3D semiclassical model.

PACS numbers: 32.80.Rm, 03.65.Sq, 73.40.Gk

While the realization that a "particle" could traverse a classically impenetrable, static potential barrier arose early in the development of quantum mechanics, there has been continuing interest in (and some controversy [1] over) quantum-mechanical tunneling. Questions being raised [1] include the definition of tunneling time(s) and how a nonstationary barrier is traversed. Several theories [2–4] address the tunneling ionization of atoms in an oscillatory electric field with amplitude F. Tunneling interpretations have been applied to some intense-laser multiphoton ionization (LMPI) experiments [5–8], often with noble gas atoms or their positive ions. However, given that tunneling rates increase exponentially with F and that absolute determination of relevant peak intensities $>10^{12}$ W/cm^2 in focused laser beam pulses is said [8,9] to be difficult even to a factor of 2, LMPI experiments have not provided *sensitive* tests of dynamic tunneling theory. Indeed, the term tunneling ionization was even ascribed to LMPI experiments that were analyzed with a model based on classical, over-the-barrier escape [6(a)]. In this Letter we use tunneling to refer specifically to a transition *through* a barrier, where the transition connects two states having the same total energy; we also contrast this with mechanisms, such as MPI, involving transitions to states with other energies.

Our data come from microwave ionization of excited H atoms. Microwave technology facilitates precise determinations [10] of the field amplitude and pulse shape, a major advantage over LMPI experiments. Moreover, the simplicity of the hydrogen atom allows us to model details of the ionization process and make a direct comparison between experiment and theories. We find that a theory [4] often used [6–8] to model laser tunneling ionization experiments fails to describe the present data.

It is convenient to use classically scaled variables [11] (Ω_0,F_0) for the frequency ω and amplitude F (atomic units are used throughout) of the applied field, where $\Omega_0=n_0^3\omega$ and $F_0=n_0^4F$. The classical dynamics [11,12] depends on only these and not separately on ω, F, and the principal classical action I_0, here set equal (in a.u.) to n_0. Using results from Ref. [13] for each parabolic substate $\mathbf{n}=(n_0,n_1,|m|)$, the highest [lowest] critical field $F_{\rm cl}^{\rm crit}(\mathbf{n})$ for classical escape is $F_0=0.38$ [$F_0=0.13$] for the extremal upward-going [downward-going] (in energy as F increases) substate $(n_0,n_0-1,0)$ [$(n_0,0,0)$]. Tunneling systematically lowers the onset of static ionization to lower F_0 values.

This Letter presents the first precise experimental and theoretical exploration of what we shall call the "dynamic tunneling regime," which, for the present conditions, is $0\leq\Omega_0\lesssim0.05$. (This is one of six regimes [14] of dynamical behavior yet identified for the microwave ionization of H atoms in various ranges of Ω_0.) The experimental apparatus has been described previously [12,15]. Briefly, H^+-Xe electron-transfer collisions produced a 17-keV beam of neutral H atoms. A fraction of about 10^{-5} was in a given parabolic state of $n_0=7$, from which $^{12}C^{16}O_2$ laser double-resonance excitation took place to an extremal parabolic state in an n_0 manifold between 24 and 32. Previous [12,14,15] and present work showed the atomic substate distribution entering the microwave cavity [16] to correspond classically to a microcanonical ensemble of initial orbits with fixed I_0.

With the slow turn-on and turn-off $A(t)$ of the microwave amplitude in the atomic rest frame [16], the system may be described [17] by the Hamiltonian $\mathcal{H}(t)=p^2/2-1/r+A(t)zF\sin(\omega t+\varphi)$; the initial phase φ of the $\omega/2\pi=9.908$ GHz microwave field was averaged by the experiment.

For each of $n_0=24,27,31$, Fig. 1 shows a typical, measured ionization curve, where ionization means true ionization plus excitation to bound states above an apparatus-determined cutoff n value, n_c [12,14,15]. Experimental insensitivity to variations of n_c values in the range 75–90, i.e., far above n_0, along with calculations, justifies our assumption that true ionization completely dominated. Each $n_0<32$ curve rose monotonically and smoothly from ionization probability $P_{\rm expt}=0\%$ to 100%. Also shown are calculations using several theoretical models discussed next.

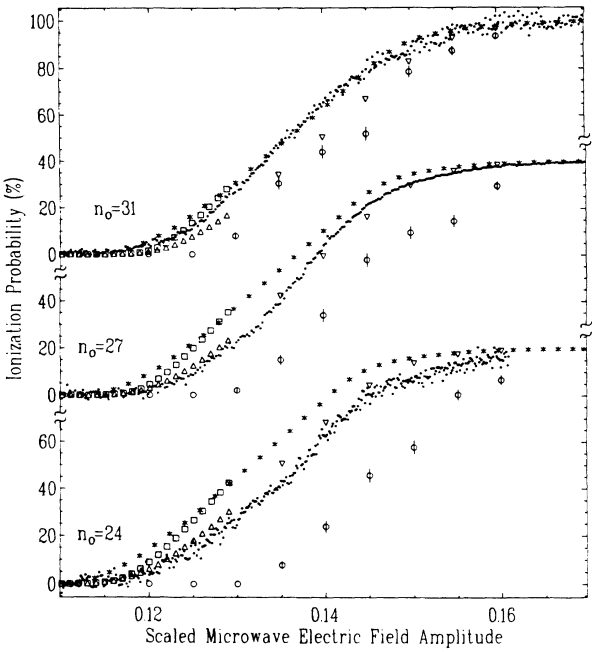

FIG. 1. ●, experimental data for $n_0=24,27,31$; □, DK-formula model calculations P_{DK}; △ (▽), semiclassical (extended) calculations P_{sc} (P_{sc}^{ext}); *, "adiabatic" model calculations P_{cl}^{adia}; ○, classical 3D Monte Carlo calculations P_{cl}^{MC} (with statistical error bars).

The "adiabatic" model assumes that (i) F_0 is large enough to mix strongly the initial n_0 and adjacent (n_0+1) manifolds; (ii) F_0 exceeds F_{cl}^{crit} for some (n_0+1) substates; (iii) these ionize completely. The adiabatic ionization probability P_{cl}^{adia} is obtained from an average of the classical critical [13] field $F_{cl}^{crit}(\mathbf{n})$ over the (n_0+1) manifold. In this limit P_{cl}^{adia} depends only upon F_0 and can be computed by Monte Carlo integration over the microcanonical substate distribution, if one allows for the influence of the alternating sign of the field on $F_{cl}^{crit}(\mathbf{n})$.

This approximation is complementary to tunneling, which assumes no transitions between adiabatic basis states. The coupling constant [18] for item (i) is $C_{n_0}=1.5n_0(F_0)(n_0^3\omega)$. For fixed ω and F_0, this varies as n_0^4. In particular, the ratio $C_{24}/C_{31}\simeq 1/3$. Notice that experimental 3D data in Fig. 1 agree fairly well with the calculated P_{cl}^{adia} (asterisks in Fig. 1) for $n_0=31$, less well for $n_0=27$, and rather poorly for $n_0=24$.

One may begin to explore dynamic tunneling by integrating the *static* tunneling rate $\Gamma_S(F;\mathbf{n})$ for each substate over one microwave field oscillation to obtain a fractional loss of population per field period $\int dt\,\Gamma_S(\mathbf{n})\equiv W(\mathbf{n})$. After N field oscillations, the ionization probability averaged over the microcanonical substate distribution is $P_{tunn}(F_0;n_0)=\langle 1-e^{-NW}\rangle$. Although accurate numerical computation of each Γ_S is possible [19], the subsequent calculations required to produce P_{tunn} make this method numerically too intensive for $n_0\gg 1$.

Therefore, as has also been done for analyzing low-frequency LMPI experiments, we exploit approximate forms for Γ_S. Equation 72 in Ref. [19] is an analytic, semiempirical [Damburg and Kolosov (DK)] formula for Γ_S. Experiments [20] with some $|m|<3$ substates of $n_0=30,40$ showed it works reasonably well for (near) extremal substates, but it can overestimate Γ_S for other states by at least an order of magnitude. The ionization probabilities $P_{DK}(F_0;n_0)$ (open squares in Fig. 1) calculated with use of the DK formula are discussed below.

We also used uniform [Jeffreys-Wentzel-Kramers-Brillouin (JWKB)] semiclassical approximations for Γ_S to calculate the ionization probabilities $P_{sc}(F_0;n_0)$ (open triangles in Fig. 1). (Reference [21] shows good agreement between JWKB and "numerically exact" values for Γ_S.) These calculations reproduce the P_{expt} curves remarkably well for $n_0\leq 29$ and $F_0\leq 0.13$. However, when $F_0>0.13$, for some substates $F_0>F_{cl}^{crit}(\mathbf{n})$. For them we set $P=1$ because their ionization occurs on a time scale significantly less than the total interaction time; we treat the other, below-the-barrier substates semiclassically. This "extended" semiclassical model gives upper bounds, P_{sc}^{ext} (open inverted triangles in Fig. 1); it should be a good approximation for our pulse length [16], but it would overestimate the ionization probability for very short pulses.

Finally, we used a classical 3D Monte Carlo model [11,12], in which Hamilton's equations are integrated for orbits randomly chosen from an initial microcanonical distribution. The resultant classical ionization probabilities P_{cl}^{MC} (open circles in Fig. 1) always underestimate the present P_{expt}, and the discrepancy increases with decreasing n_0. [For the lower values of n_0 (alternatively, Ω_0), the discrepancy near the onset of ionization is quantitatively similar to that for classical versus tunneling ionization in a static field. For example, fields measured in Ref. [20] to produce static ionization rates in the range $\Gamma_S=10^5$-10^8 s^{-1} for individual substates of $n_0=30,40$ are from 8% to 17% smaller than corresponding classical values [13] of $F_{cl}^{crit}(\mathbf{n})$.] However, in Fig. 1 the disagreement of both P_{cl}^{MC} and P_{cl}^{adia} with P_{expt} seems to decrease as F_0 rises. We infer a growing importance of dynamic couplings between bound states as F_0 increases.

We may now exploit the precise calibration [10] of the microwave amplitude to see that our data distinguish finely between the different theoretical ionization mechanisms, in particular tunneling versus over-the-barrier escape. All our data, e.g., those in Fig. 1, have the same 5% absolute amplitude uncertainty but the *relative* amplitude uncertainty between data for different values of n_0 is even smaller. One may clearly distinguish for $n_0=24,27$ in Fig. 1 between the semiclassical (extended) model(s) and DK formula model calculations as the latter model yields curves having a shape different from the experimental curves. A small global adjustment of the experimental amplitude does not change this trend. Both models involve tunneling, but the DK-formula mod-

el is inferior for these lower n_0 values at least in part because the DK formula overestimates Γ_S.

For higher n_0 values, e.g., $n_0=31$ in Fig. 1, the apparent agreement between the DK-formula model and experiment is accidental. As Ω_0 increases, C_{n_0} increases, causing the initial n_0 manifold to become significantly coupled to the (n_0+1) manifold, with a consequent increase in the ionization probability. Hence, at some Ω_0 (or n_0), the experimental microwave ionization curve will accidentally lie near the DK-formula curve.

Especially for $P_{\text{expt}} \gtrsim 20\%$, the adiabatic model calculations reproduce very well the $n_0=31$ experimental curve but not the curves for lower n_0 values. The ionization mechanism assumed for this model, strong n_0 to (n_0+1) excitation followed by classical escape, is very different from tunneling.

Figure 2 compares experimental $F_{\text{expt}}(10\%)$ (solid circles) and calculated $F_0(10\%)$ values, $F_0(X\%)$ being the microwave amplitude at which $X\%$ ionization occurs. Within the error bars showing measurement reproducibility, the measured 10% values are *not flat* versus n_0. (A comparison of weighted, least-squares fits with a polynomial function with orders up to 0,1,2, respectively, strongly supports this assertion.) The average $\langle F_{\text{expt}}(10\%)\rangle$ is near 0.124; the average 50% value $\langle F_{\text{expt}}(50\%)\rangle$ is near 0.135 (see, e.g., Fig. 1). These data show that a $1/9n_0^4$ scaling law, claimed on the basis of data for 50% microwave ionization probability for Li and Na "hydrogenic" ($|m|=2$) Rydberg states [22], is not quantitatively correct for H atoms at low Ω_0.

The 3D classical Monte Carlo calculations (open circles) only start to approach $F_{\text{expt}}(10\%)$ for the higher n_0 values in Fig. 2. At the lower end, not only is the $\gtrsim 10\%$ disagreement significant, but the trend is wrong: With decreasing n_0 the calculations rise whereas the measurements gently fall.

Results of the adiabatic model (asterisks) are in near agreement with $F_{\text{expt}}(10\%)$ only for n_0 between about 29 and 32, which shows that in a 9.9-GHz field the ionization mechanism in this model is wrong for lower n_0 values.

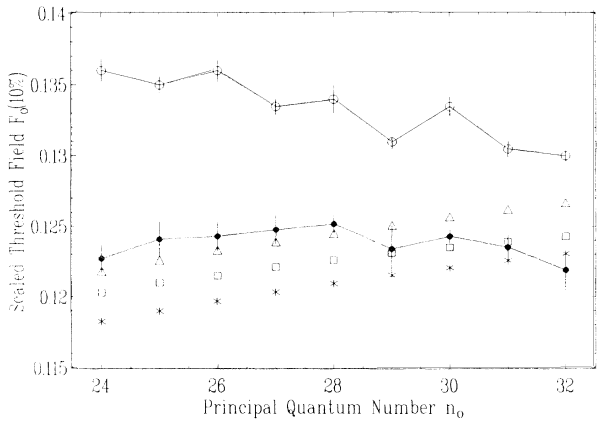

FIG. 2. $F_0(10\%)$ thresholds as a function of n_0 (symbols same as in Fig. 1). The experimental error bars show the reproducibility for each n_0. Not shown is an overall 5% uncertainty in absolute experimental field strength.

In contrast, the semiclassical tunneling theory (open triangles) agrees best with $F_{\text{expt}}(10\%)$ only at the lower end, $n_0 \simeq 24$–29. Though close, the DK-formula model calculations (open squares) drop below $F_{\text{expt}}(10\%)$ for $n_0=24$–28. Note that both calculations decrease monotonically with decreasing n_0 in Fig. 2, whereas the experimental results peak mildly near $n_0=28$, right where the ionization mechanism changes.

For H atoms exposed to about 3×10^2 oscillations of a 9.92-GHz field, the comparisons in Figs. 1 and 2 show tunneling to become the dominant ionization mechanism for $n_0 \leq 28$, or $\Omega_0 \leq 0.033$. Notice that for an initial $n_0=28$ atom the classical electron hits the moving barrier $(2\Omega_0)^{-1}=15$ times per half period of the oscillatory field, which sets a time scale for tunneling to become important. Earlier studies [12,14,15] showed the importance of coupled-state ionization mechanism(s) at higher Ω_0; even classical calculations gave good estimates of ionizing field amplitudes for $\Omega_0 \gtrsim 0.1$, or $n_0 \gtrsim 40$ at 9.9 GHz.

We continue with quantal models [3,4] inspired by Keldysh [2], who initiated attempts to describe oscillatory-field ionization for a range of frequencies from the tunneling limit ($\gamma \ll 1$) to the MPI limit ($\gamma \gg 1$); the "Keldysh parameter" γ is the ratio of the tunneling time (see the second sentence of this Letter) and the field period. Though these *double* inequalities are consistent with Keldysh's original presentation [2], one finds in the LMPI literature [6(a)] $\gamma < 1$ (not $\gamma \ll 1$) being called "the tunneling regime." For H atoms $\gamma=\omega/n_0F=n_0^3\omega/n_0^4F=\Omega_0/F_0$ [13]. With γ ranging from 0.40 down to 0.17 for our 10%-ionization fields, $\gamma < 1$ is an insufficient condition for tunneling.

Moreover, our data do not support another published tunneling condition, viz., that of Ref. [23]: "...the tunnel condition for highly excited states ($n^* \gg 1$) is $\gamma^2 n^* \ll 1$." Our present data cover the range $0.69 \leq \gamma^2 n_0 \leq 5.2$. At $n_0=28$, where we observe tunneling to set in for a 9.9-GHz field, $\gamma^2 n_0=2.0$.

Keldysh or later theories have been used to model recent LMPI experiments [5–8]. Using Eq. 9.24 of Ref. [4(b)], which is the Ammosov-Delone-Krainov (ADK) model applied to H atoms and which should be valid for arbitrary initial states [24] in the tunneling regime, we calculated $F_0(10\%)$ for a microcanonical substate distribution. The ADK values (not shown) are nearly a factor of 2 below experimental values for $n_0=24$–32. Since a tunneling ionization rate depends exponentially on F, this is a *huge* discrepancy. Alternatively, evaluating ADK at experimental $F_{\text{expt}}(10\%)$ values gives ionization probabilities above 97%, far too high.

We have found the main reason for this failure. For $n_0 \gg 1$ the tunneling rate formula [4] is a product of a prefactor and a factor of the form $\exp[-n_0 g(F_0;n_0)]$, where $g(F_0;n_0)$ varies relatively slowly with n_0. When

$n_0 \gg 1$, g depends only on F_0 and the ratios n_1/n_0, n_2/n_0, and $|m|/n_0$. The amplitude at which $g=0$ is clearly important: For large n_0 this is just $F_{cl}^{crit}(\mathbf{n})$. *The exponential factor in ADK [4] has its zero at the wrong value of F.* For $n_0 \simeq 1$ this leads to small errors. Our calculations for $n_0=1$ show that the three values of $F_0(10\%)$ obtained from ADK, the DK-formula model, and the present semiclassical method all agree. However, the error in ADK rises dramatically with increasing n_0. Therefore, the assertion (Ref. [7], p. 863) that "...the validity of the ADK theory improves as n^* increases" is wrong for Rydberg atoms.

In summary, our low-scaled-frequency microwave ionization data, with precision much beyond that of pulsed-laser experiments, distinguish finely between different theoretical models near and past the onset of dynamic tunneling. Whereas only Keldysh-type theory was available years ago for interpreting the first H-atom microwave ionization data [25], and it is still often applied to pulsed-laser experiments, our experimental and theoretical results clearly reveal its limitations. The extended model introduced in this Letter yields the first 3D quantum-mechanical calculations to reproduce microwave ionization data for 3D excited hydrogen atoms.

We thank the National Science Foundation and Open University for support.

---

[a]Present address: Department of Physics and Astronomy, University of Wyoming, Laramie, WY 82071.

[1] E. H. Hauge and J. A. Støvneng, Rev. Mod. Phys. **61**, 917 (1989).

[2] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].

[3] A. M. Perelomov, V. S. Popov, and V. P. Kunznetsov, Zh. Eksp. Teor. Fiz. **54**, 841 (1968) [Sov. Phys. JETP **27**, 451 (1968)]; F. H. M. Faisal, J. Phys. B **6**, L89 (1973); H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).

[4] (a) M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)], which unfortunately contains misprints and errors. Corrected versions of Eqs. 21 and 22 agree with Eq. 9.24 for H atoms in (b) N. B. Delone and V. P. Krainov, *Atoms in Strong Light Fields* (Springer, Berlin, 1985).

[5] K. G. H. Baldwin and B. W. Boreham, J. Appl. Phys. **52**, 2627 (1980); F. Yergeau, S. L. Chin, and P. Lavigne, J. Phys. B **20**, 723 (1987); S. L. Chin, C. Rolland, P. B. Corkum, and P. Kelly, Phys. Rev. Lett. **61**, 153 (1988); M. D. Perry, A. Szöke, O. L. Landen, and E. M. Campbell, Phys. Rev. Lett. **60**, 1270 (1988).

[6] (a) S. Augst, D. Strickland, D. D. Meyerhofer, S. L. Chin, and J. H. Eberly, Phys. Rev. Lett. **63**, 2212 (1989); (b) W. Xiong and S. L. Chin, Zh. Eksp. Teor. Fiz. **99**, 481 (1991) [Sov. Phys. JETP **72**, 268 (1991)].

[7] S. Augst, D. D. Meyerhofer, D. Strickland, and S. L. Chin, J. Opt. Soc. Am. B **8**, 858 (1991).

[8] G. Gibson, T. S. Luk, and C. K. Rhodes, Phys. Rev. A **41**, 5049 (1990).

[9] G. Petite, P. Agostini, and F. Yergeau, J. Opt. Soc. Am. B **4**, 765 (1987).

[10] B. E. Sauer, K. A. H. van Leeuwen, A. Mortazawi-M, and P. M. Koch, Rev. Sci. Instrum. **62**, 189 (1991).

[11] J. G. Leopold and I. C. Percival, Phys. Rev. Lett. **41**, 944 (1978).

[12] D. Richards, J. G. Leopold, P. M. Koch, E. J. Galvez, K. A. H. van Leeuwen, L. Moorman, B. E. Sauer, and R. V. Jensen, J. Phys. B **22**, 1307 (1989).

[13] D. Banks and J. G. Leopold, J. Phys. B **11**, 37 (1978).

[14] P. M. Koch, L. Moorman, and B. E. Sauer, Comments At. Mol. Phys. **25**, 165 (1990).

[15] P. M. Koch, L. Moorman, B. E. Sauer, E. J. Galvez, K. A. H. van Leeuwen, and D. Richards, Phys. Scr. **T26**, 51 (1989).

[16] The 4.96-cm-long, 5.32-cm-diam cylindrical cavity resonated cw in the TM_{020} mode at 9.908 GHz. The 0.63-cm-thick end caps had 0.26-cm-diam holes on axis to pass the 0.09-cm-diam beam. The field distribution was calculated numerically after H. Halbach and R. Holsinger, Part. Accel. **7**, 213 (1976). In the atom rest frame the pulse $A(t)$ turned on and off in 22 cycles (5% to 95%) and was constant ($>95\%$) over 250 cycles.

[17] J. G. Leopold and D. Richards, J. Phys. B **24**, 1209 (1991).

[18] D. Richards, J. Phys. B **20**, 2171 (1987).

[19] R. J. Damburg and V. V. Kolosov, in *Rydberg States of Atoms and Molecules,* edited by R. F. Stebbings and F. B. Dunning (Cambridge Univ. Press, New York, 1983).

[20] P. M. Koch and D. R. Mariani, Phys. Rev. Lett. **46**, 1275 (1981).

[21] J. A. C. Gallas, H. Walther, and E. Werner, Phys. Rev. Lett. **49**, 867 (1982).

[22] T. F. Gallagher, C. R. Mahon, P. Pillet, P. Fu, and J. B. Newman, Phys. Rev. A **39**, 4545 (1989).

[23] N. B. Delone and V. P. Krainov, J. Opt. Soc. Am. B **8**, 1207 (1991).

[24] Note that Keldysh-type ionization formulas that assume an atom initially in an H(1*s*)-like state describe our data very poorly.

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