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Microwave Ionization of Hydrogen Atoms: Experiment versus Classical Dynamics

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Ionization of hydrogen atoms with principal quantum number $n=32$, 40, and 51–74 by a 9.92-GHz electric field $\mathbf{F}(t)=\hat{\mathbf{z}}F_0\cos\omega t$ was studied with a superimposed static electric field $\bar{F}_s=0$, 2, 5, and 8 V/cm. The measured field strengths $F_0(10\%)$ at which 10% of the atoms were ionized are in excellent agreement with classical calculations in both one and two spatial dimensions. Covering finer detail as well as gross structure of the n dependence of $F_0(10\%)$, the agreement supports the application of classical dynamics to the analysis of this strongly perturbed quantum system.

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Early experiments on the microwave ionization of highly excited hydrogen atoms[1] stimulated subsequent theoretical work.[2] Classical Monte Carlo calculations of electron trajectories[3] furnished the first few theoretical data in agreement with experiment.[4]

Here we present measured threshold fields $F_0(10\%)$ at which 10% of a hydrogen atom beam prepared in a given n manifold was ionized by 9.92-GHz microwaves and make a detailed comparison with new classical calculations in one and two spatial dimensions that were performed using methods described by Jensen[5] and Leopold and co-workers.[6–8] The agreement is striking over nearly the complete range, $n=32$, 40, and 51–74, whose extremes were determined by present experimental conditions. This establishes that classical dynamics, wherein the threshold for ionzation corresponds to the onset of chaotic motion in the classical nonlinear system, provides a useful description of a real, strongly perturbed quantum system. Furthermore, the n dependence of the experimental $F_0(10\%)$ values reflects the presence of periodic orbits and surrounding island structures[5,6] in the classical phase space near scaled microwave frequencies $n^3\omega=\frac{1}{4}$, $\frac{1}{3}$, $\frac{2}{5}$, and $\frac{1}{2}$.

We do not imply that quantal calculations should not be pursued for this manifestly quantal system. Quite the contrary; but quantal calculations of the microwave ionization of excited hydrogen are difficult because in a strong external field huge numbers of bound levels and the continuum must be included. The first quantal calculations[9] for a one-dimensional (1D) hydrogen atom used a strictly bound basis of up to 200 states and led to a conclusion that the chaotic diffusion upward in energy, which constitutes the classical mechanism for ionization, would be limited in the quantum system. In light of the excellent agreement displayed in this Letter, however, statements that the agreement between the early experiments[1] and classical calculations[3] was accidental[10] may have been premature.

Recent experiments[11] elsewhere emphasized the study of n-changing transitions of hydrogen atoms prepared in extremal (quasi one-dimensional) Stark states in combined microwave and static electric fields. Quantal calculations[12] on a discrete basis qualitatively agreed with the experimental n-changing results. Experimental curves[11] labeled "ionization," however, exhibited thresholds which have been noted[13] to be about half of those obtained with 1D classical calculations.

The method used in the present experiment is described elsewhere.[14] Electron-transfer collisions of an $\simeq 14$-keV proton beam in an Xe gas cell produced an n^3-weighted beam of fast hydrogen atoms. A double-resonance method employing two CO_2 lasers excited those in the $(n, n_1, |m|) = (7, 0, 0)$ extremal Stark state, via the $(10, 0, 0)$ state, to a selected $(n, 0, 0)$ state. Because the laser beams crossed the atomic beam at shallow angles,[14] they did not enter the microwave interaction region.

About 0.5 m downstrem from the laser-excitation regions, the atoms traversed a cylindrical Cu cavity of 4.96-cm length and 2.66-cm radius, operated in the TM_{020} mode. Its microwave electric field axis $\hat{\mathbf{z}}$ and the atomic beam axis coincided. Microwave fringe fields extended over about one to two times the $\simeq 7$-mm diameter of the holes in the entrance and exit end caps. Thus, each 14-keV atom experienced about 300 microwave oscillations at constant amplitude F_0 between an adiabatic rise and fall of $F(t)$ over about 40–80 oscillations. For most of the measurements, a 0.07-mm-thick annular Mylar spacer isolated each end cap from the cavity body.[15] This enabled application of a potential difference across the end caps to superimpose with $F(t)$ a collinear static electric field $\bar{F}_s$.[16]

Because of variations in the strength and direction of electric and stray magnetic fields after the laser-excitation region, the substate distribution of the atoms was altered before they entered the cavity. Field ionization[17] in a set of static electric field plates located about 15 cm before the cavity was used to diagnose the distribution entering it at four sampled values, $n = 56$, 61, 66, and 70. A comparison with Monte Carlo calculations based on Eq. (6) of Damburg and Kolosov[18] indicated consistency with equally populated substates and unchanged n.

A static voltage V_{label} applied to the cavity body enabled "energy-labeled" detection[19] with a particle multiplier of protons produced inside the cavity. However, the resultant static electric field F_{label} after the cavity ionized any atoms excited by microwaves into $n \gtrsim 81$ bound states. If the ionization occurred close after the cavity, the protons were approximately energy-labeled and detected, albeit with a reduced efficiency. Therefore, the experimental signal consisted of microwave ionization plus excitation to $n \gtrsim 81$ states. Ionization curves registering the microwave power dependence of this signal for each n value were recorded at $\bar{F}_s = 0$, 2, 5, and 8 V/cm.

It is interesting to compare in Fig. 1 ionization curves for different pairs of neighboring n values. Those for $n = 67$ and $n = 68$ at each value of $\bar{F}_s$ are nearly identical and do not vary too much with $\bar{F}_s$. Contrast this with those for $n = 61$ and $n = 62$, which differ greatly at the lower $\bar{F}_s$ values, but at $\bar{F}_s = 8$ V/cm have become nearly identical, with a much gentler slope.

To effect a comparison with calculations based on classical dynamics, we extracted from each curve an $F_0(10\%)$ value. They are close to the apparent onsets of ionization because the curves rise rapidly. Furthermore, with one-dimensional orbits being expected to be among those most easily ionized classically,[20] comparison of $F_0(10\%)$ values with 1D calculations might be warranted. Figure 2 displays experimental $F_0(10\%)$ values for $n = 51–74$ at the four $\bar{F}_s$ values. There is a distinct staircaselike behavior in each curve. It may be

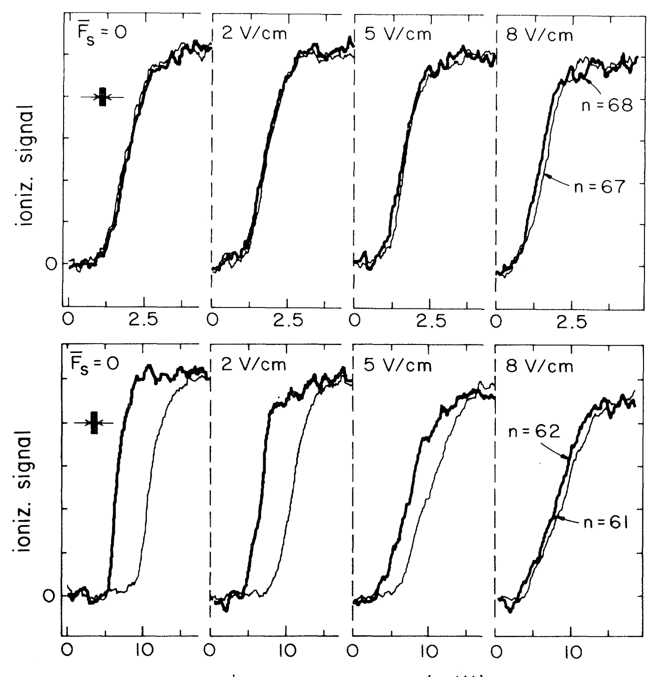

FIG. 1. Signal-averaged microwave ionization curves (smoothed over the horizontal segments shown) as a function of the power incident on the microwave cavity. Each frame shows curves for two adjacent n values taken with superimposed static electric field $\bar{F}_s = 0$, 2, 5, and 8 V/cm.

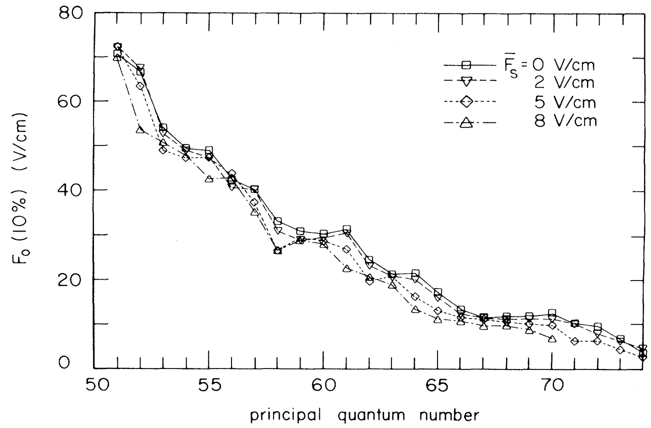

FIG. 2. Microwave field strengths $F_0(10\%)$ (on the axis of the cavity) at which 10% of hydrogen atoms prepared in a given n level were ionized while traversing the cavity, at $\bar{F}_s = 0$, 2, 5, and 8 V/cm; see Refs. 16 and 21.

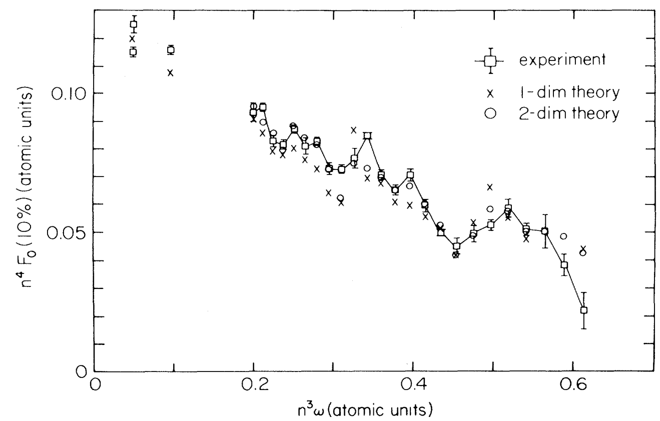

FIG. 3. Classically scaled 10% threshold fields $[n^4F_0(10\%)]$ vs the classically scaled microwave frequency (n^3): A comparison for $\bar{F}_s = 0$ of experimental and 1D and 2D classical calculations. See Refs. 16, 21, and 22.

interesting to note that increasing $\bar{F}_s$ seems to cause a displacement of a given curve in Fig. 2 toward lower n.

The classical calculations were carried out with the 1D surface-state-electron (SSE) model[5] and the 2D drogen atom model,[7,8] using the Monte Carlo method described in Refs. 3 and 6. Each included important aspects of the experimental situation; the presence of $\bar{F}_s$, the number of oscillations of the microwave field, and its adiabatic turnon and turnoff. Figure 3 compares the experiment with the 1D and 2D calculations for the case $\bar{F}_s = 0$. The axes correspond (vertical) to the threshold field $n^4F_0(10\%)$ classically scaled to the Coulomb binding field and (horizontal) to the microwave frequency $n^3\omega$ classically scaled to the unperturbed classical orbital frequency. At low scaled frequencies the data approach the threshold for ionization through (quantal: $n^4F \simeq 0.12$ a.u.[17,18]) or over (classical: $n^4F \simeq 0.13$ a.u.[23]) the potential barrier in a Coulomb plus static field. The general decrease with increasing scaled frequency, punctuated with localized structure, indicates the importance of the dynamics.

The two nonoverlapping experimental points at $n^3\omega \simeq 0.05$ $(n = 32)$ correspond to data taken with (upper point) and without (lower point) Mylar-isolated end caps.[15] The discrepancy might have been due to heating of the Mylar spacers in the relatively high (up to 8 W) microwave power needed to ionize $n = 32$, causing a minute displacement of the cavity end caps and a resultant variation in the cavity Q or coupling factor K.[14] Therefore, we believe that the lower point is preferable. No such discrepancies were observed at higher n values, where the microwave power levels were significantly lower.

The agreement between experimental and calculated $F_0(10\%)$ values is remarkable, particularly when one compares the structures in the curves. In the classical nonlinear dynamics, the structures reflect resonances[5,6] at, e.g., $n^3\omega = \frac{1}{4}, \frac{1}{3}, \frac{2}{5}$, and $\frac{1}{2}$; there, relatively more stable orbits lead to higher thresholds for the onset of chaos.

That the 2D results are generally significantly closer than the 1D ones to the experiment is not too surprising: The experimental substate distribution corresponds classically to a microcanonical ensemble[3] of orbits in three spatial dimensions. However, the discrepancies between both calculations are not large with the 1D SSE model obviously giving a good, but usually low, estimate of the 10% ionization thresholds.

Experiment and available 2D calculations differ mildly at $n = 52$ and $n = 69$, which are near classical resonances at $n^3\omega = \frac{1}{5}$ and $\frac{1}{2}$, respectively; they differ significantly at $n = 59$ and $n = 61$, which straddle the resonance at $n^3\omega = \frac{1}{3}$. The range $n \geqslant 73$ challenges both the experiment, which faces increasing sensitivity to $\bar{F}_s$, F_{label}, and stray fields and decreasing signal-to-noise ratio, and the theory, which must include the effects of F_{label}. For $n \leqslant 72$, agreement also persists for $\bar{F}_s \neq 0$. The shift of the curves in Fig. 2 toward lower n and F_0 as $\bar{F}_s$ increases is well reproduced by both 1D and 2D calculations (not shown).

As a step toward explaining the apparent discrepancy between our results and the curves labeled "ionization" in Ref. 11, it may help to point out the main differences between the two experiments. In Ref. 11 the collinear excitation laser beam was present in the microwave interaction region. Because different atoms were laser excited in the microwave field and throughout the waveguide (into quasi one-dimensional states), the ensemble did not experience a given number of total oscillations; the maximal number was nearly an order of magnitude larger than in the present experiment. The observed[11] quantal multiphonon processes[12] may have played a dominant role during the longer time scale of Ref. 11.

In summary, the present work reveals excellent agreement between experimental ionization threshold fields and those calculated using classical dynamics. The classical mechanism responsible for ionization is the onset of chaotic motion of the electron in the combined Coulomb microwave field. The agreement extends over a wide range of initial n values and includes the effect of a static electric field on the ionization threshold.

An important remaining question is exactly what is the range of conditions under which the classical dynamics remains useful for the description of a manifestly quantal system. For the microwave ionization of hydrogen, a detailed answer awaits further experimental and theoretical efforts. The challenge remains to quantum theory to explain the experimental results in this strongly perturbed regime.

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[15]The isolated and nonisolated microwave configurations were separately calibrated with comparable accuracy.

[16]F_s was calculated on axis to be uniform to $\pm 25\%$ about a central value $\bar{F}_s$, with a z variation similar to one oscillation of a cosine.

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[20]However, calculations found some 2D thresholds below 1D ones, e.g., $n = 60$, 68, and 69 in Fig. 3.

[21]Individual relative errors (90% confidence) for $n \leq 71$ are $\simeq 3\%$ and reflect spread in the data. For $n > 71$ they increase. The microwave calibration procedure adds $\pm 5\%$ uncertainty in the absolute F_0 scale. At the radial edge of the atomic beam F_0 was about 8% lower than on axis; see Ref. 14.

[22]An adiabatic (versus sudden) turnon, especially, was found to be important in the calculations, but variation over the estimated experimental range of 40–80 oscillations changed the $F_0(10\%)$ values by less than the estimated theoretical (Monte Carlo) errors. In Fig. 3, for the 1D calculations, these are ± 1 V/cm, except for $n = 32$ and 40, which are $< \pm 2\%$. For the 2D calculations, these are $< \pm 2\%$, except for $n = 59$, 68, and 71, which are $< \pm 3\%$.

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