Prediction of a New Peak in Two-Frequency Microwave "Ionization" of Excited Hydrogen Atoms

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(Received 28 June 1993)

We report the first theoretical and experimental study of the detailed frequency dependence of the "ionization" of excited H atoms by a two-frequency ($2f$) microwave field. Our 1D quantal and 1D classical calculations, in excellent agreement with each other, predict a new $2f$ ionization peak. The peak scales classically. We present a model which shows that the location and the width of the peak are determined by classical phase-space structures. The theoretical results are supported by experimental data with 3D atoms.

PACS numbers: 05.45.+b, 32.80.Rm, 42.50.Hz

Ionization of an atom by a pulse of intense electromagnetic field whose photon energy is much less than the atomic binding energy attracts both experimental and theoretical attention [1–4]. Even for one-frequency ($1f$) driving, theory is challenged by the number of important parameters. (i) For the field: the pulse envelope function $A_1(t)$, peak electric amplitude F_1 [3], angular frequency $\omega_1 = 2\pi\nu_1$, and polarization $\hat{\mathbf{e}}_1$; (ii) for the atom: the quantum numbers of the initial, (near-) resonant intermediate, and final (continuum) state(s). This Letter addresses the even more interesting case of two-frequency ($2f$) driving, which adds parameter subscripted 2 to those subscripted 1 in (i). By restricting our attention to fixed ω_2 and F_2, identical pulse shapes $A_1(t) = A_2(t) = A(t)$, and linear polarization $\hat{\mathbf{e}}_1 = \hat{\mathbf{e}}_2$, we present the first study of the detailed frequency (ν_1) dependence of the $2f$ "ionization" probability P_{ion} of H atoms with large principal quantum number n_0. Ionization denotes true ionization plus excitation to final bound states above an experimentally specified n cutoff $n_c^q > n_0$ [5]. They key point of this Letter is to predict theoretically and to support with experimental data a prominent nonmonotonic frequency-dependent structure in P_{ion}.

Hydrogen, the simplest atom, is most amenable to accurate theoretical modeling and numerical simulations. Because of the high principal quantum numbers involved in microwave-driven H($n_0 \gg 1$) atoms, one expects that even classical theoretical methods can provide insight into the ionization dynamics [5–7].

The most powerful way to establish a link between quantal and classical dynamics in the driven H atom is to demonstrate [6] classical scaling of measured or calculated quantities, which now discuss for $2f$ driving. Ignoring negligible spin and relativistic effects, in atomic units a.u. ($e = \hbar = \mu = 1$, μ being the reduced electron mass) the Hamiltonian is $\mathcal{H}(t) = \mathbf{p}^2/2 - |\mathbf{r}|^{-1} + z\,A(t)[F_1 \times \sin(\omega_1 t') + F_2 \sin(\omega_2 t' + \delta\varphi)]$, where $A(t) = \sin(\pi t/T)$, $0 \le t \le T$, and $t' = t + t_0$. The variation in the time t_0 at which each atom enters the microwave field causes phase averaging, whereas the relative phase $\delta\varphi$ between the two microwave modes can be fixed, e.g., by phase locking two microwave sources.

A similarity transformation [6] shows that the classical dynamics depends only on the classically scaled frequencies $\Omega_i \equiv n_0^3\omega_i$ (the ratio of ω_i and the initial Kepler or Bohr orbital frequency $1/n_0^3$) and the scaled amplitudes $\epsilon_i \equiv n_0^4 F_i$ (the ratio of F_i and the Coulomb field binding the electron to the Bohr orbit with principal quantum number $n = n_0$). Thus, the classical dynamics does not depend separately on n_0, ω_i, and F_i. As noted in Ref. [6] for $1f$ driving, the quantum dynamics is different because the similarity transformation does not preserve the canonical commutation relations; we interpret $[\tilde{x}, \tilde{p}_x] = i\hbar/n_0$ as defining an effective $\hbar$, $\tilde{\hbar} = \hbar/n_0$.

We now present the main result of this Letter, prediction of a new ionization peak (and minima that flank it) for excited H atoms driven by a $2f$ field, along with the first supporting experimental data. Moreover, our calculations show that for at least a 23% variation in $\tilde{\hbar}$ (n_0 ranging from 57 to 70), not only does the quantal $2f$ ionization peak scale classically, there is also quantitative agreement among 1D quantal calculations, 1D classical calculations, and a mapping approximation to the latter. We expect this $2f$ ionization phenomenon will stimulate additional theoretical and experimental work. This happened earlier for $1f$ ionization of H atoms: Theoretical discovery of "a new photoelectric peak" [8] led to a flurry of theoretical publications on the classical and quantal dynamics of this chaotic system; other nonmonotonic structures, such as "subthreshold resonances" [9, 10] and "scars" [6, 11, 12] also spurred both experiment and theory.

Consider the dynamics of the 1D H atom defined by $\mathcal{H}(t)$ (with $r = z > 0$). This Hamiltonian is known to be an excellent approximation for describing the dynamics of those hydrogen states which are most easily ionized [4, 7, 9, 11, 13]. We solved numerically the

0031-9007/94/73(2)/248(4)$06.00

Schrödinger equation $i\partial_t|\psi(t)\rangle = \mathcal{H}(t)|\psi(t)\rangle$, $|\psi(t=0)\rangle = |n_0\rangle$, in an unperturbed basis $n \in [50, 220]$. Guided by the accessible range of parameters in the experiment to be described below, we chose $F_1 = 14.5$ V/cm, $F_2 = 8.2$ V/cm, $\nu_2 = 34.998$ GHz, $n_0 = 57$, $n_c^q = 86$, and $T = 4.3$ ns for the envelope function $A(t) = \sin(\pi t/T)$. We defined $P_{\text{ion}} = \sum_{n=n_c^q+1}^{220} |\langle n|\psi(t=T)\rangle|^2$. Figure 1 shows P_{ion} for ν_1 between 9 and 45 GHz. The quantal results in Fig. 1 clearly reveal a resonance structure. This by itself would not be too surprising because, due to the discrete level structure of the H atom, one should expect resonance structures here and there. The exciting news is that *1D classical calculations also quantitatively reproduce the resonance structure.* The classical results were obtained by numerically integrating the classical equations of motion for 200 trajectories whose initial conditions were chosen to model the quantum state $|n_0\rangle$ as closely as possible.

This quantal-classical correspondence naturally suggests investigating whether the quantal calculations follow classical scaling [6]. This is indeed so. Figure 2 shows how for $n_0 = 57$ and 70 the 1D quantal (and, for comparison, 1D classical) P_{ion} varies with scaled frequency Ω_1. With ν_k, F_k, T, and n_c^q denoting the values given above for $n_0 = 57$, the $n_0 = 70$ calculations used the classically rescaled values $\tilde{\nu}_k = (57/70)^3\nu_k$, $\tilde{F}_k = (57/70)^4 F_k$, $\tilde{T} = (70/57)^3 T$, and $\tilde{n}_c^q = 1 +$ [integer part of $(70/57)n_c^q$]. The quantitative agreement among the scaled quantal and classical calculations strongly suggests that the "resonance" behavior in the frequency dependence of P_{ion} can be explained classically. This observation together with Howard's idea of interspersed resonances [14] led us to a simple model for describing the ionization process.

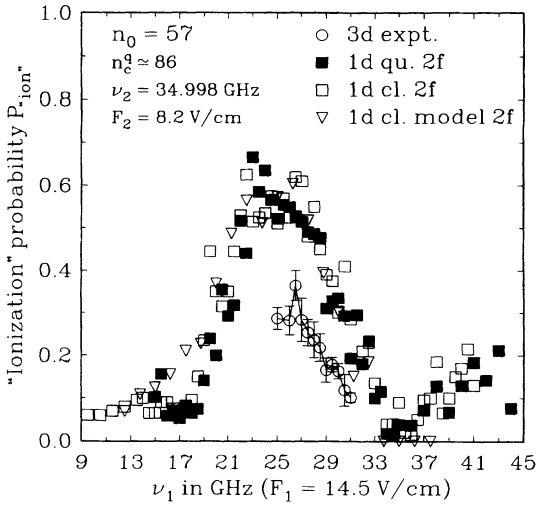

FIG. 1. Frequency (ν_1) dependence of experimental (3D), numerical (quantal and classical 1D), and model (classical 1D) $2f$ "ionization" probability P_{ion} for fixed ν_2, F_1, and F_2.

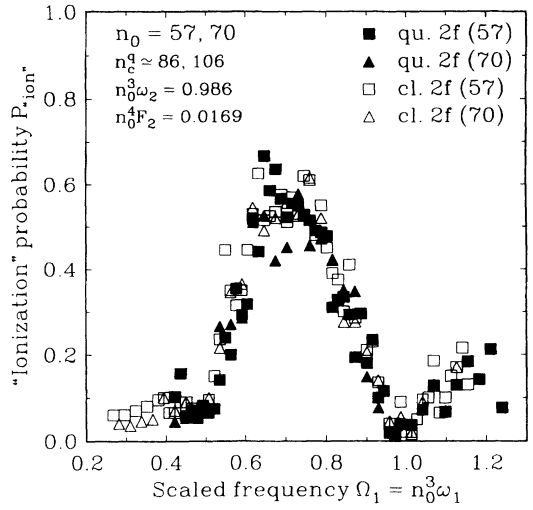

FIG. 2. Demonstration of classical scaling of 1D quantal and 1D classical $2f$ calculations ($n_0^4 F_1 = 0.0298$).

The ionization mechanism is best explained in (θ, I) phase space, which is obtained from the z, p_z representation by a canonical transformation $p_z = \cot[\eta(\theta)]/I$, $z = 2I^2 \sin^2[\eta(\theta)]$, $\theta = 2\eta - \sin(2\eta)$. The action variable I is the classical analog of the quantal bound state η value. The bound-motion dynamics governed by $\mathcal{H}(t)$ can be visualized in the (θ, I) plane by choosing several initial conditions (θ_0^j, I_0^j), $j = 1, 2, \ldots, N$ and marking them as well as their M iterates after time $t_m = m\Delta t$, $m = 1, \ldots, M$ by a dot. For $\nu_1 = 25$ GHz, $\nu_2 = 35$ GHz, $\Delta t = 8.26 \times 10^6$ a.u., $N = 30$, and $M = 30$ the result is shown in Fig. 3. In Fig. 3 four "resonance structures" can be seen that are caused by either ω_1 or ω_2 being in resonance with $\omega = I^{-3}$ a.u. The resonance condition is $\omega_i I^3 = r_i/s_i$ where r_i and s_i are integers. One sees that for $r_2 = s_2 = 1$ the structure at $I_1 \approx 57.3$ as well as the two structures at $I_2 \approx 72.2$ ($r_2 = 2$, $s_2 = 1$) are due to the driving frequency ω_2 while the interspersed resonance at $I_I \approx 64.1$ ($r_1 = 1$, $s_1 = 1$) is due to ω_1.

Now consider the case of an arbitrarily chosen frequency ν_1. For ionization to occur it is necessary that the interspersed resonance [14] overlaps [15] with *both* the $I_2 \approx 72.2$ and $I_1 \approx 57.3$ resonances. The position of the interspersed resonance is given by $I_I = 187.35/\nu_1^{1/3}$ where ν_1 is in GHz. Using a formula from [14] for the resonance width, we obtain the following two conditions for resonance overlap: (i) $68.8[A(t)F_1]^{1/2}/\nu_1 + 2.0[A(t)F_2]^{1/2} > I_I - I_1$, for the I_1 and I_I resonances; and (ii) $68.8 \times [A(t)F_1]^{1/2}/\nu_1 + 2.2[A(t)F_2]^{1/2} > I_2 - I_I$, for the I_I and I_2 resonances. F_1 and F_2 are in V/cm. When (i) and (ii) are fulfilled simultaneously during a time interval $\tau(\nu_1) > 0$, ionization may occur [e.g., for $\nu_1 = 25$ GHz, $\nu_2 = 35$ GHz, and $A(t) = 1$ the conditions are already fulfilled $F_1 \approx 3.4$ V/cm and $F_2 \approx 1.9$ V/cm]. During $\tau(\nu_1)$ we assume decay of the phase-space probability (ionization)

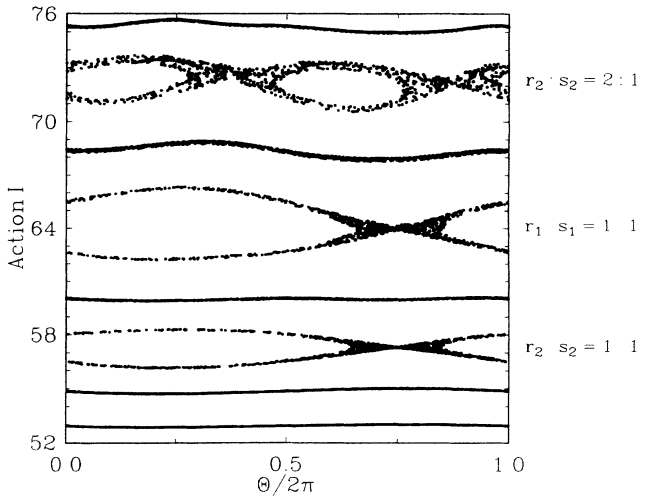

FIG. 3. Phase-space structure of $2f$ driven 1D hydrogen atoms. Parameters of $\mathcal{H}(t):A(t) = 1$, $\delta\varphi = 0$, $t_0 = 0$, $\nu_1 = 25$ GHz, $F_1 = 0.725$ V/cm, $\nu_2 = 35$ GHz, $F_2 = 0.41$ V/cm.

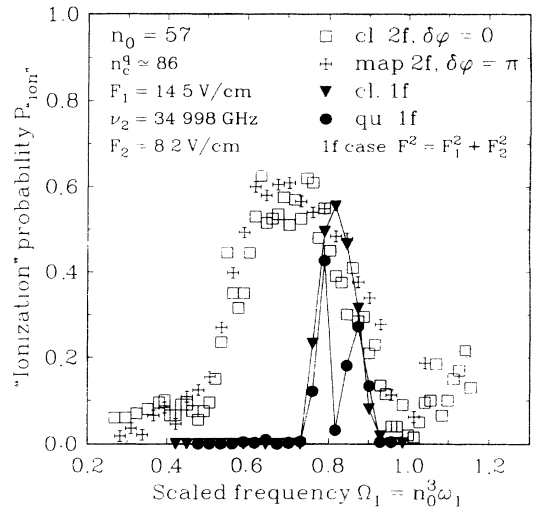

FIG. 4. Ionization effectiveness of $1f$ vs. $2f$ driven H atoms exposed to the same microwave power. 1D classical $2f$ calculations ($t_0 = 0$): numerical integration of Hamilton's equations for $\delta\varphi = 0$; iterated symplectic map $\mathcal{M}$ showing little difference in P_{ion} for $\delta\varphi = \pi$.

with an average rate $\lambda(\nu_1)$. The model ionization probability is then given by $P_{\text{ion}}^{(M)}(\nu_1) = 1 - \exp[-\lambda(\nu_1)\tau(\nu_1)]$. We approximated $\lambda(\nu_1)$ by $\frac{1}{3}$ of the decay constant obtained numerically for $A(t) = 1$. The result for $P_{\text{ion}}^{(M)}$ is shown in Fig. 1. The sharp cutoff of $P_{\text{ion}}^{(M)}$ for $\nu_1 > 33$ GHz is due to $\tau(\nu_1) = 0$ because the interspersed resonance no longer overlaps with the $I_2 \approx 72.2$ islands for the values F_1, F_2, ν_2 in Fig. 1. The center of the P_{ion} peak occurs when $\tau(\nu_1)$ is maximal at $\nu_1 = 25$ GHz. For $\nu_1 < 25$ GHz, the decrease in $\tau(\nu_1)$ causes the decrease on the left wing of the P_{ion} peak. Details of this analysis will be reported elsewhere [16].

Figure 4 shows calculations for $n_0 = 57$ addressing the interesting question of how much more effective for ionization a $2f$ field is compared to a $1f$ field. The 1D quantal (basis $n \in [50, 200]$) and classical (400 trajectories) $1f$ calculations used $F(t) = F\sin(\pi t/T)\sin(\omega_1 t)$, $0 \le t \le T$. We chose $F^2 = F_1^2 + F_2^2$ to give the same $1f$ power as in the $2f$ case, where $F_1 = 14.5$ V/cm and $F_2 = 8.2$ V/cm. Notice the 1D classical $1f$ ionization peak that is much narrower than the 1D classical $2f$ peak, thought the drop of each near $\Omega_1 = 0.9$ is comparable. However, the 1D quantal $1f$ calculations show a sharp dip in P_{ion} for Ω_1 just above 0.8. Based on recent experimental and theoretical work [12] that found the same effect, we conjecture that the nonclassical $1f$ local stability at the dip is the consequence of a scarred wave function [12, 17].

The method used to generate Fig. 3 can also be interpreted as a mapping $\mathcal{M}$ which generates the dot (θ_m, I_m) from (θ_{m-1}, I_{m-1}). Extending the approach reported in [18] we arrived at an explicit analytical representation of $\mathcal{M}$ [16]. Figure 4 shows that the results of our $2f$ map quantitatively agree with our 1D classical $2f$ numerical integration calculations.

The mapping $\mathcal{M}$ permitted efficient investigation of the dependence of P_{ion} on phase changes resulting from varying t_0 and $\delta\varphi$. Calculations confirmed our expectation of only very weak variation with t_0 as long as $T \gg (\nu_2 - \nu_1)^{-1}$, a condition fulfilled in the experiment we describe below. We found the same behavior for variations in $\delta\varphi$. Thus, the "ionization peak" is robust against changes in the initial phase $\delta\varphi$. Moreover, exploratory calculations with the $2f$ map showed that the classical local stability (i.e., local minima in P_{ion}) near $\Omega_1 = 0.5$ and 1 repeats for Ω_1 near 2 but not near 1.5.

We now present experimental evidence supporting our theoretical results. In the apparatus described in [12, 19], we produced a 14.6 keV beam of $H(n_0)$ atoms that entered a Ka band (26–40 GHz) Cu waveguide interaction region in a uniform distribution of $n_0 = 57$ substates (classically a microcanonical ensemble). 0.53 mm diam holes centered in each short sidewall allowed the beam to traverse a $2f$ field in the waveguide, whose spatial TE_{10} mode produced (in the atomic rest fame) the $A(t) = \sin(\pi t/T)$, $0 \le t \le T = 4.3$ ns pulse shape used in the theoretical calculations above. The output of a Gigatronics model 900 synthesizer was amplified with a Miteq model AFD 4-080180-2P preamplifier and frequency doubled with a Honeywell model A2000N doubler to produce the variable frequency ν_1. The 11.666 GHz output of a Gigatronics model 910 synthesizer was frequency tripled with a Honeywell model A2300N tripler producing the fixed frequency $\nu_2 = 34.998$ GHz. Using 10 MHz clock pulses from the model 900, the two syn-

thesizers were phase locked. The $2f$ field was combined in a directional coupler, amplified with a 1 W Hughes model 1077H12F00 26–40 GHz traveling wave tube amplifier (TWTA), and led into the vacuum system through a vacuum window to cross the atomic beam. It was finally dumped in a well matched (voltage-standing-wave ratio ≤ 1.0001) absorbing load inserted directly into the waveguide inside the vacuum. We determined the field amplitudes $F_1 = 14.5$ V/cm and $F_2 = 8.2$ V/cm with 5% uncertainty and kept the TWTA shot noise level [12] below $\simeq 0.8$ V/cm rms. Excited atoms not "ionized" by the $2f$ field were ionized further downstream in a longitudinal static field ionizer. The resulting ions were detected with a Johnston MM1 particle multiplier whose signal pulses were processed and stored by a computer system. We interpret the experimental quench curves (not shown) as recording microwave ionization with an n cutoff $n_c^q \simeq 86$ [5].

The resulting experimental data for P_{ion} as a function of ν_1 are shown in Fig. 1. They can be interpreted as the high-frequency wing and the top of the resonance structure predicted by our theoretical calculations. Unfortunately, the low-frequency cutoff of our microwave system prevented us from obtaining experimental data below 25 GHz. That these data were obtained for fixed $F_1 + F_2$, however, demonstrates that an amplitude-sum rule of thumb for determining approximate $2f$ ionization threshold amplitudes [20] is not always applicable. Exceptions to this rule were already noted in Refs. [4, 20, 21].

The $2f$ quantal calculations shown in Fig. 1 are a factor of 2 higher than the experimental data. This accords with previous $1f$ [9–11] and $2f$ [13] 1D quantal calculations that overestimated P_{ion} compared to measurements with 3D H atoms but still reproduced well the variation of P_{ion} with F or ν.

In summary, we obtained the following results: (i) Based on detailed 1D quantum and classical calculations, we predict the occurrence of a $2f$ ionization peak that scales classically. (ii) Based on a picture of frequency dependent classical phase-space structures, we present a physical model that explains the occurrence, the location, and the width of the ionization peak. (iii) Our experimental data support the theoretical predictions.

Including both experiment and calculations, this first investigation of the detailed frequency dependence shows nonmonotonic behavior in $2f$ ionization of excited H atoms that calls for further experimental and theoretical study.

We thank J. Howard for lively discussions and suggestions in the initial stages of this project. Work at Stony Brook is supported by the NSF. R. B. appreciates financial support by the Deutche Forschungsgemeinschaft and a generous grant from the University of Delaware Research Foundation.

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