Polarization Independence of Microwave "Ionization" Thresholds of Excited Hydrogen Atoms near the Principal Resonance

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(Received 19 January 1995)

We study experimentally and theoretically how the polarization of a 9.904 GHz field affects threshold amplitudes for many-photon "ionization" of $n_0 = 70, \ldots, 98$ H atoms. That peak-field thresholds for circular polarization are *lower* than for linear polarization (with those for elliptical polarization intermediate) in the main resonance zone is a consequence of a time-scale separation in the 3D dynamics there. When an amplitude scaling relationship given by the theory is used, the experimental data confirm that, near onset, the scaled ionization thresholds are approximately independent of polarization.

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

The polarization of an intense electromagnetic field can strongly influence atomic ionization when many photons must be absorbed. In a perturbation expansion polarization-dependent electric dipole selection rules determine pathways through unperturbed states, so small changes in polarization may dramatically vary the ionization rate for fixed peak-field amplitude. Except for the case [1] of few-photon ($\lesssim 3$) ionization dominated by a resonance between intermediate bound states, early optical [2] and microwave [3] experiments, using atoms in initial states $|n_0, \ell, m\rangle$ with low values of the angular momentum ($\ell \leq 2$) and a driving frequency $\omega \ll (E_{n_0+1} - E_{n_0})/\hbar$, showed that many-photon ($\gtrsim 11$) ionization by a circularly polarized (CP) field required significantly larger peak-field amplitudes than a linearly polarized (LP) field. A smaller effective quantal density of participating, unperturbed states in CP fields was used to explain [4] these many-photon results: The monotonic change of the azimuthal quantum number m with each absorbed photon forces an eventual monotonic increase in ℓ, whereas in LP the $\Delta m = 0$ selection rule does not seriously constrain the upward and downward movement in ℓ permitted by the $\Delta\ell = \pm 1$ selection rule. Classically, quasistatic pictures including effects of frame rotation and an angular momentum barrier were used [5,6] to explain the microwave ionization results. Nevertheless, these explanations are all "rules of thumb," applicable only to particular cases. They have no universal validity. In a succinct description of our limited knowledge a recent review [7] states, "the matter of polarization . . . is more complicated than previously realized."

All CP experiments and most theories to date have focused on fields with frequency ω less than the atomic Kepler frequency ω_K. (The energy $\hbar\omega_K$ is near the mean of the $\Delta n = \pm 1$ unperturbed energy splittings.) When $\omega/\omega_K \ll 1$, a quasistatic view should be applicable if narrow quantal resonances [8] are avoided. At higher frequencies dynamics should play an important role. Classically, an approximate resonance analysis led to a conclusion that the onset of ionization for H atoms driven at $\omega \simeq \omega_K$ by a CP field would occur at a peak amplitude estimated to be 1.5–2 times below that for an LP field [9].

Unlike the LP and CP cases, the elliptically polarized (EP) case has no integrals of the motion: It has three degrees of freedom and is not conservative. Moreover, the case that interests us here, that of very many unperturbed states being strongly coupled, is unlikely to be amenable to any simple selection rule analysis for LP vs CP vs EP driving. To tackle this difficult problem we consider frequencies at which the dynamics of H atoms in LP fields is dominated by the main classical resonance, at which the scaled frequency $\Omega_0 \equiv \omega/\omega_K \simeq 1$, so the LP dynamics can be locally approximated by a 1D pendulumlike Hamiltonian that elucidates much of the physics. Here we demonstrate how this resonance dynamics, extended to the 3D case, also allows one to understand the similarity of ionization of atoms by CP and EP fields when $\Omega_0 \simeq 1$.

In our experiment we used established techniques [10] to prepare, via two-step laser excitation, a 14.6 keV beam of H atoms in a uniform distribution of substates with given principal quantum number n_0 in the range $70, \ldots, 98$. The beam was collimated to 0.21 cm before entering the 9.904 GHz microwave field inside a TE_{121} mode cylindrical brass cavity [1]. Atoms that were not "ionized" by the microwave field, i.e., neither ionized nor excited to final bound states above an n cutoff [10,11], $n_c^q \simeq 110$ (determined by a 3.83 V/cm static electric field that was entirely outside the cavity, the stray static field inside the cavity being at least 10^2 times smaller), were detected downstream via microwave ionization in a voltage-labeled TE_{101} mode rectilinear cavity. We interpret the measured survival probability as $1 - P_{\text{ion}}$, where P_{ion} is the "ionization" probability [10,11].

Microwave power entered the cavity via two off-axis slits in the entrance end cap, allowing

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excitation of two spatially orthogonal, frequency-degenerate, LP modes to create the on-axis field, $\mathbf{F}(t) = \lambda(t)\mathcal{F}(\hat{x}\alpha\sin(\omega t) + \hat{y}\sin(\omega t + \delta))$, where $\lambda(t)$ describes the half-sine, 153 cycle, microwave pulse envelope experienced by the atoms traversing the cavity. Note that the ratio $F/\mathcal{F}$ of the peak amplitude F to the amplitude $\mathcal{F}$ of the $\hat{y}$ component depends on polarization through the parameters α and δ, where $0 \le \alpha \le 1$ and $0 \le \delta \le 90°$. The output of a synthesized source [12(a)] passed through a switch [12(b)] was amplified [12(c)] and split [12(d)] approximately equally into two arms. Before entering the vacuum system and the cavity, the microwaves could be attenuated [12(e)] in one arm and phase shifted [12(f)] in the other arm. With the vacuum system open to air we used a polarization sensitive detector to sample the cavity field leaking out the exit end-cap hole so that we could adjust tuning screws in the side walls to minimize the frequency splitting (170 kHz) and cross coupling of the two modes [13]. Setting the relative amplitude and phase of the two modes to $\alpha = 1$ and $\delta = 90°$, respectively, would achieve CP. In practice we could use the atoms to fine-tune to CP [14]; this gave $\alpha = 1.00 \pm 0.05$ and $\delta = 90° \pm 1°$. We created EP, while keeping F constant, by reducing α to 0.263 ± 0.016 via a variable attenuator [12(e)]; we kept $\delta = 90°$ by adjusting the phase shifter [12(f)] to null a measured 1° phase shift introduced by the attenuation. Alternatively, we could have set $\alpha = 1$ and varied δ, in which case the peak amplitude would become a function of δ, but the field *intensity* (photons/sec m^2) would remain fixed. We created LP by extinguishing the power in one arm ($\alpha \le 0.0002$).

Using the classically scaled [10,15] amplitude $F_0 = n_0^4 F$ and frequency $\Omega_0 = n_0^3\omega$ (unless otherwise noted, we use atomic units) and $F_0(X)$ for the peak-field amplitude at which $P_{\text{ion}} = X$; Fig. 1(a) [1(b)] presents the $F_0(0.1)$ [$F_0(0.5)$] data. The experimental error bars indicate the highest and lowest values we ever obtained for that point from several (3–8) ionization curves taken for each value of n_0 and polarization; symbols are at each mean value. That the LP thresholds are systematically a factor of 1.1 to 2 above previous 9.9 GHz LP data [10] must be a consequence of both the higher n_c^q and shorter interaction times in the present experiment. However, the similar dependences of F_0 vs Ω_0 in Figs. 1(a) and 1(b) imply that the qualitative effect of the resonance island on the mean dynamics is independent of polarization. But note that for the values of Ω_0 shown all CP $F_0(0.1)$ thresholds in Fig. 1(a) [and nearly all CP $F_0(0.5)$ thresholds in Fig. 1(b)] lie *below* corresponding LP thresholds, with EP thresholds lying between them. To our knowledge this is the first experimental observation of the *enhancement* of many-photon ionization (i.e., lower thresholds) by CP. Here the minimum number of photons needed to excite the atoms from n_0 to above n_c^q ranges from about 41 for $n_0 = 70$ to about 8 for $n_0 = 98$.

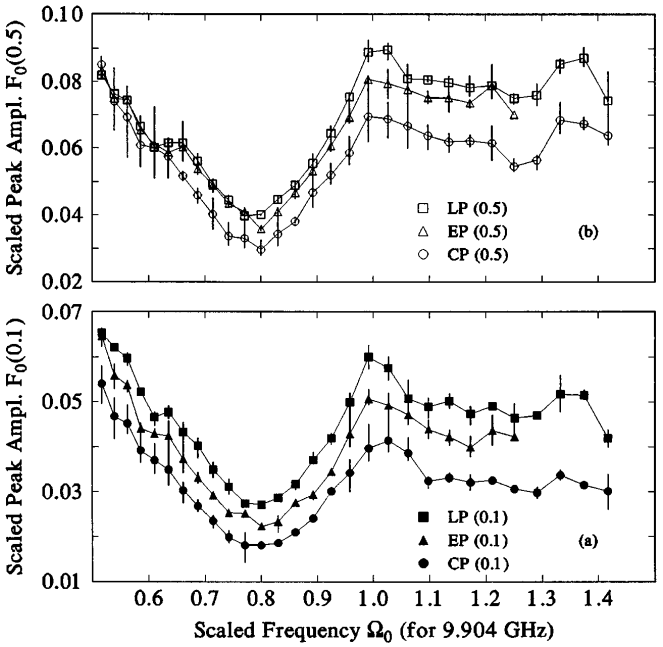

FIG. 1. Frequency and polarization dependence of experimental "ionization" *peak-field* thresholds (filled symbols, $P_{\text{ion}} = 0.1$; open symbols, $P_{\text{ion}} = 0.5$) for H($n_0 = 70, \ldots, 98$) atoms.

Figure 2 compares the $F_0(0.1)$ and $F_0(0.5)$ data with 3D classical Monte Carlo (3D CL) calculations for CP, EP, and LP that modeled [16] all important features of the experiment, including the uniform distribution of initial substates, the pulse envelope, $\lambda(t)$, and the n cutoff, n_c^q. Note the relatively good agreement of the data with the classical simulations for all polarizations. There are also local disagreements that have been explained previously for LP [17–21], especially the enhanced, nonclassical, local stability of the data over 3D CL simulations near $\Omega_0 = 0.82$ and 1.35. Indeed, Ref. [21] used supercomputer calculations at $\Omega_0 = 1.304$ to demonstrate numerically the near "eternal" lifetime of such (separatrix) electronic wave packets for excited 3D H atoms driven by an LP field. The present CP and EP data, particularly the $F_0(0.5)$ data in Figs. 1(b), 2(a), and 2(b), also seem to exhibit (though a bit less strongly) the nonclassical local stability shown previously for LP [17–20] to be caused by quantal separatrix states of the main resonance island. That this phenomenon seems to be unchanged by variations in the polarization of the strong driving field is an important new result calling for 3D quantal calculations [21–23] and theory.

In Fig. 3 we replot the experimental data from Fig. 1, using a field parametrized with amplitude $\mathcal{F}$ for $\alpha = 1$ and variable δ. For our [LP] {EP} (CP) data, this $\mathcal{F}$ is $[1/\sqrt{2}]$ $\{1/1.26\}$ (1.00) times the peak amplitude F. The plots of the classically scaled thresholds $\mathcal{F}_0 \equiv n_0^4\mathcal{F}(X)$ vs Ω_0 in Fig. 3 are *insensitive to polarization near the main resonance for low X*; e.g., for $\Omega_0 > 0.63$ the $\mathcal{F}_0(0.1)$ thresholds are nearly identical for LP, EP, and

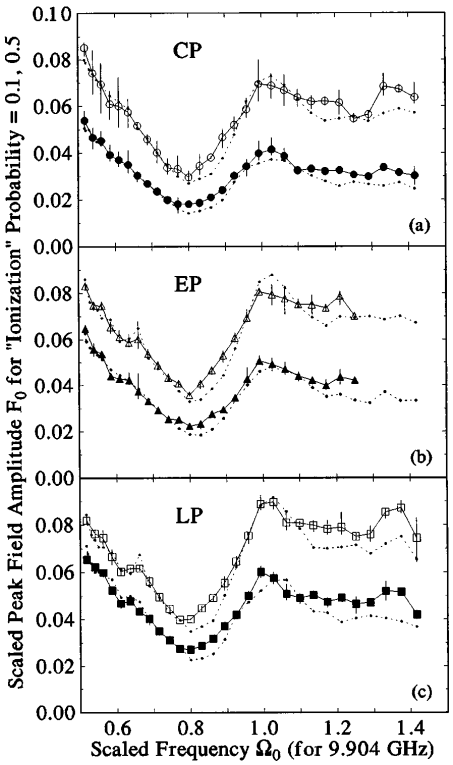

FIG. 2. The same data shown in Fig. 1 compared with the results of 3D classical Monte Carlo simulations (dotted lines) for (a) CP, (b) EP, (c) LP.

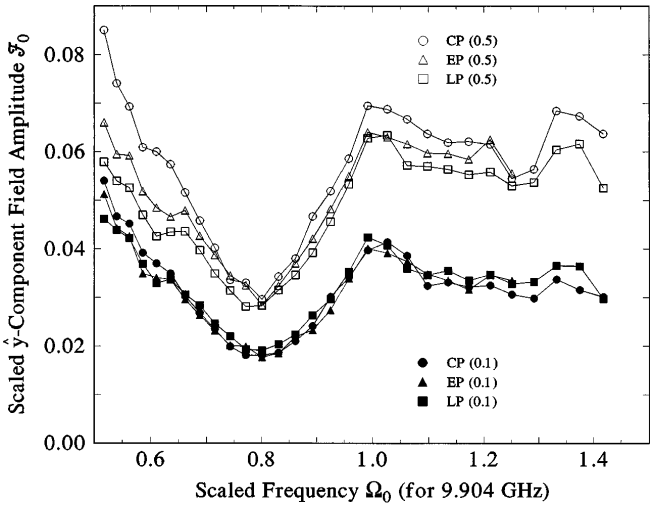

FIG. 3. Frequency and polarization dependence of the measured ionization thresholds expressed in terms of the scaled $\hat{y}$-*component* field amplitude $\mathcal{F}_0 = n_0^4\mathcal{F}$ defined in the text.

CP. *However, this agreement persists only up to* $X = 0.3$; *at* $X \gtrsim 0.5$ *the CP thresholds are systematically higher.*

Encouraged by the agreement of the data with classical simulations, and realizing that the high values of n_0 and inherent 3D nature of this problem strain the most advanced quantal computations [21–23], we have developed a classical theory to explain our results; Ref. [16] will give all details. Here we provide only a very brief qualitative explanation of why classical dynamics leads to the invariance of $\mathcal{F}_0(X)$ with respect to polarization, observed for $X \leq 0.3$, and how the main resonance causes it. The dynamics of the 3D hydrogen atom is determined by the Hamiltonian $H = p^2/2 - 1/r + \mathbf{r}\cdot\mathbf{F}(t)$, where $\mathbf{F}(t)$ was given earlier. On using the unperturbed, $\mathbf{F} = 0$, angle-action variables $(\theta_n, \theta_l, \theta_m, I_n, I_l, I_m)$ [24], we can show that near the principal resonance, where $\omega = \omega_K(I_n)$, because $\dot{\theta}_n \simeq \omega_K$ the system has three distinctly different dynamical time scales: Fastest is the field variation, slowest is the changing orientation of the Kepler ellipse, and intermediate is the change in θ_n. If follows (see [16]) that for $\omega \simeq \omega_K$ there is a canonical representation in which the Hamiltonian has the form $K = K_p + K_f$, where K_f is a rapidly varying part that causes the escape from the regular, slow motion due to K_p.

The slow Hamiltonian, K_p, may be approximated by one similar to that of a plane vertical pendulum subject to gravity, with a "gravitational" coefficient that is a function of the slowly varying angle-action variables,

$$K_p = -\frac{3p^2}{2I_R^2} + \lambda(t)\mathcal{F}_0 H_r(\Theta;\delta)\cos q\,,$$

where $\Theta = (\theta_l, \theta_m, I_l, I_m)$, I_R is the value of the principal action satisfying $\omega = \omega_K(I_R)$, and (q, p) are simply related to the variables describing the motion around the Kepler ellipse. Reference [25] analyzes motion produced by a Hamiltonian similar to K_p, and Ref. [24] shows that H_r is an approximate adiabatic invariant and that the variables Θ vary more slowly than (q, p) which, in turn, vary more slowly than K_f. Moreover, H_r is relatively weakly dependent upon δ, the field ellipticity; the mean of H_r^2 over a microcanonical distribution is independent of δ.

The slowly varying field envelope adds the final, and slowest, time scale that is important to the ionization dynamics [26]. Including its effect we can, e.g., estimate the ionization threshold at the minimum of $\mathcal{F}_0(X)$ near $\Omega_0 = 0.8$. For $\Omega_0 < 1$ the first orbits to ionize as $\mathcal{F}_0$ increases satisfy two conditions [19]: (i) They adiabatically switch onto the separatrix of K_p as $\lambda(t)$ slowly increases from 0 to 1. (ii) The amplitude $\mathcal{F}_0$ must be large enough for the first ($\Omega_0 = 1$) and second ($\Omega_0 = 2$) resonance islands to touch: this is the Chirikov overlap criterion [27]. When (i) is satisfied, the initial phase points gradually move onto the separatrix of K_p and then wind around the edge of the resonance island. The presence of K_f converts separatrices into stable and unstable manifolds. When (ii) is satisfied, a proportion of orbits transfers to the unstable manifold of the $\Omega_0 = 2$ island and subsequently to higher actions that easily ionize.

These two conditions provide estimates for the frequency of minimum threshold, near $\Omega_0 = 0.8$, and also the critical values of $\mathcal{F}_0$ at which ionization first occurs,

$\mathcal{F}_0^c(\Theta, \delta)$; Ref. [16] will give details. Since the variables Θ are slowly varying functions of time, the minimum of this function gives an estimate of the critical field. We find that this minimum is only weakly dependent upon δ. As δ increases from 0 to $\pi/2$, $\min[\mathcal{F}_0^c(\Theta, \delta)]$ decreases monotonically from about 0.014 to 0.011 and Ω_0^c from 0.78 to 0.77. In the 3D CL simulations the minimum of $\mathcal{F}_0(0.1)$ decreases from 0.016 to 0.014, a statistically insignificant change, and $\Omega_0^c \simeq 0.8$, for all δ.

In summary, our analysis using the approximate pendulumlike Hamiltonian K_p gives a theoretical explanation for the insensitivity to polarization displayed in Fig. 3 of $\mathcal{F}_0(X = 0.1)$ ionization thresholds. We close with two conclusions: (i) We deduce that the pendulumlike resonance is much more important in governing the strong-field dynamics of the atom than is the cumulative effect of polarization-dependent selection rules that determine each of the steps along the pathway to ionization. Because such resonances are generic features of mixed-phase-space classical systems and have a clear quantal analog [19,20,28,29], this result should not be regarded as a special case. It is interesting to ask what determines the boundaries of the influence of this resonance. We tentatively conclude that the marked divergence of the thresholds for different polarization in Fig. 3 at $\Omega_0 \simeq 0.63$ is the signature of its lower boundary, but analysis to demonstrate this theoretically must be left for future work. (ii) At higher scaled frequencies, $\Omega_0 > 2$, classical resonance analysis leads us to believe that the classical $\mathcal{F}_0(X)$ thresholds are independent of the polarization for low X; this has been confirmed by 3D CL calculations [16], but bearing in mind the significant differences between classical and quantal LP dynamics when $\Omega_0 > 2$ [11,30,31], only further work will confirm that this remains true for the quantal case.

We are grateful to the NSF, EPSRC, and Schlumberger-Doll Research for financial support of this work, to A. Birnbaum for help in its early phase, and to T. Gallagher for discussions and sharing results of work in advance of publication.

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