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INTERACTIONS OF INTENSE MICROWAVES WITH RYDBERG ATOMS

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Résumé.— Nous présentons une revue d'expériences qui s'intéressent à l'ionisation, l'excitation, et la structure quantique des atomes d'hydrogène et d'hélium très excités plongés dans un champ de micro-onde intense, ainsi qu'une comparaison avec les théories actuelles. Celles-ci sont capables d'expliquer qualitativement certaines structures étudiées, mais elles ne permettent pas encore d'expliquer quantitativement les mesures d'ionisation effectuées à l'aide d'un très grand nombre (plusieurs centaines) de photons micro-onde. Il existe des calculs par la méthode de Monte-Carlo basés sur une théorie classique des interactions qui donnent un accord raisonnable avec les expériences d'ionisation publiées. Ces calculs négligent l'effet tunnel (quantique) et des expériences sont actuellement en cours pour montrer si cet effet, négligé dans l'approche classique, joue un rôle important. D'autres expériences récentes montrent que la dépendance de l'ionisation en fonction de la puissance micro-onde d'un atome de Rydberg d'hélium dans l'état triplet $m=0$ est très différente de celle d'un atome d'hydrogène. On est capable actuellement d'expliquer le seuil d'ionisation de l'hélium en termes d'interactions dynamiques aux croisements évités successifs du diagramme de potentiel Stark de l'hélium.

Abstract. — Experiments investigating ionization, excitation, and quantal structure of highly-excited hydrogen and helium atoms in an intense microwave field are reviewed in terms of existing theory. Quantal theory is able to explain some of the results of the structure studies but is not yet at the level of producing quantitative agreement with ionization experiments, which involve very large numbers (hundreds) of microwave photons. Published Monte-Carlo calculations based on a classical theory of the interactions are in reasonable agreement with the results of published ionization experiments, but additional experiments are being carried out to see if quantal tunneling, which is ignored by the classical theories, plays an important role. New experiments show that the power dependence of microwave ionization of $m=0$ triplet helium Rydberg atoms is very different from that of hydrogen atoms. We are able to explain the threshold region of ionization in helium in terms of dynamic interactions at the first (and subsequent) avoided crossings(s) of adiabatic helium Stark potential curves.

1. **Introduction.**— This paper reviews briefly our present experimental and theoretical understanding about what happens when an atom with one of its electrons in a highly-excited level with principal quantum number $n \gg 1$ (to be called a Rydberg atom) interacts with an intense microwave electromagnetic field. We shall see that much remains to be understood about this problem, both qualitatively and quantitatively. We shall confine the discussion primarily to the one-electron hydrogen atom, the subject of nearly all the still rather meager amount of theory that has been done, and to a lesser extent, to the two-electron helium atom, the simplest multi-electron atom. We shall see that hydrogen and helium atoms prepared in Rydberg states with very nearly the same energy are ionized rather differently by the microwave field and shall explain, at least partially, why this is so.

Other review articles by the present author^{1,2} and by Bayfield³⁻⁵ have covered some of the material presented here. The latter articles⁴⁻⁵, especially, reviewed

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a large body of the quantal and semi-quantal theories treating the interactions of atoms and molecules with intense electromagnetic fields. This material will not be reviewed in detail here; the interested reader should consult these earlier reviews.

A few, loose definitions are in order at the outset. We shall take "intense" to mean that the Rydberg atom undergoes multiphoton absorption and emission in the microwave field that can be of rather high order, as much as several hundred. This usually means that induced transitions to continuum states, multiphoton ionization, can complete successfully with those to lower- or higher-lying bound states, multiphoton (de-)excitation; spontaneous transitions generally play a negligible role. Viewed semi-classically, the oscillatory microwave electric field $F_0 \cos \omega t$ has a peak strength F_0 comparable to the mean Coulomb field binding the Rydberg electron to its ionic core.

The full problem may be sub-divided into the questions of (1) atomic "structure", or what are the quantized energy levels of the atom in the microwave field and (2) atomic "transitions", or what are the detailed electronic motions (in a classical sense) among the various atomic states, particularly to the continuum? We shall motivate the conclusion that, to date, quantal or semi-quantal theories have dealt better with various aspects of the structure question but less well quantitatively with the question of ionization (bound-free transitions) than have classical theories, which obviously have nothing to say about (quantal) structure but seem capable of dealing well with some aspects of the non-linear dynamics. Here there are exciting unanswered questions concerning possible "stochastic" ionization mechanisms⁶⁻¹¹. Of course, any purely classical theory ignores quantal tunneling processes, which play an important role in ionization of atoms by a static electric field. The importance of tunneling in ionization of atoms by oscillatory fields, however, is not yet well understood. Neither is the onset of stochasticity in quantal non-linear dynamical systems.

2. A "Reasonable Approximate Hamiltonian for Hydrogen. - Consider a hydrogen atom with an infinitely massive nucleus. For highly-excited levels $H(\text{high } n)$ with $n \gg 1$, the binding energy of the Bohr levels scales as n^{-2} , whereas the spacing between neighboring n -manifolds and the fine-structure and hyperfine-structure within a given n -manifold each scale as n^{-3} , albeit with diminishing constants of proportionality. Quantum-electrodynamic splittings (Lamb shifts, etc.) decrease with n even more rapidly. In the rest of this paper, we ignore fine-, hyperfine-, and QED-contributions to the energies of hydrogen levels. They are much smaller than the Stark interaction energy associated with the oscillatory electric field in an intense microwave field. (In helium, however, we shall include, via a quantum defect formulation, the electrostatic fine-structure because it greatly modifies the atomic Stark structure, particularly for levels with low values of the magnetic quantum number, $|m|=0,1$. The $|m|=0$ Stark structure is especially non-hydrogenic. For the most part, the magnetic fine-structure can still be ignored.)

At a given point in free space an applied microwave field consists of an oscillatory electric field $\vec{F}(t) = \vec{F}_0 \cos \omega t$ and an oscillatory magnetic field $\vec{B}(t) = \vec{B}_0 \cos \omega t$. For $H(\text{high } n)$ atoms, it is not hard to show that the Zeeman interaction energies with $\vec{B}(t)$ are generally much smaller than the Stark interaction energies with $\vec{F}(t)$. For a plane-wave microwave field, one can show that the ratio of the maximum Zeeman energy W_m associated with the electronic orbital magnetic moment and the maximum Stark energy W_e is of order

$$W_m/W_e \approx \alpha/2n, \quad (1)$$

where $\alpha = (137)^{-1}$ is the fine structure constant. For $n \sim 27$, this ratio is $\sim 10^{-4}$. The Zeeman energy associated with the electronic spin-angular magnetic moment is even smaller.

If the microwave field is contained in a waveguide or cavity, its electric and magnetic components will have spatial dependences which depend on the particular mode being excited. There will exist regions where one field is much larger than the other. The experiments discussed in this paper have been performed only in waveguides or cavities in which the interaction of the atoms with $\vec{F}(t)$ was totally dominant over that with $\vec{B}(t)$.

Thus it is reasonable to adopt as a Hamiltonian (in au)

$$H(\vec{r}, t) = \vec{p}^2/2 - r^{-1} + zF_0 \cos \omega t, \quad (2)$$

which includes only the non-relativistic kinetic energy of the electron and its electric interactions with the point nucleus and with $\vec{F}(t)$. This has been the starting point for all the non-relativistic theories treating hydrogen.

3. Static Electric Field Ionization. - As a prelude to describing the intense microwave experiments, it is useful to review briefly the results of our experiments at Yale on static electric field ionization of H(high n) atoms in resolved substates described by parabolic quantum numbers $(n, n_1, n_2, |m|)$. Both kinds of experiments used similar fast-beam methods; in fact, microwave ionization in a TM_{020} -mode cavity was used as a "black-box," 100% efficient detector of H(high-n) atoms in the static ionization experiments.

Fig. 1a - A schematic diagram (top view) of the apparatus used for static and microwave ionization experiments with fast beams of hydrogen or helium atoms produced by ion-atom electron-transfer collisions. The various pairs of electrodes labelled F_1 - F_5 produce static electric fields through which the atoms fly. Two continuous CO_2 laser beams may be directed either collinearly with the atomic beam or to cross it in F_1 and F_3 , respectively, where they drive transitions between Stark substates of different n -manifolds. A microwave cavity (or waveguide, not shown) is used to study microwave multiphoton ionization and excitation processes. Experimental analog signals are digitized and signal-averaged in a multichannel scaler.

Fig. 1a,b shows a schematic diagram of our apparatus. An H^+ or He^+ ion beam with laboratory kinetic energy $T \approx 3$ -20 keV undergoes electron-transfer collisions with target gas atoms, producing Rydberg atoms with approximately an n^{-3} distribution and an unknown distribution of substates. With use of various combinations of static electric fields F_1 - F_5 , continuous laser beams (argon ion or CO_2), and techniques reviewed in Ref. 2, a beam of atoms could be prepared in an n -band (a narrowed distribution of n -manifolds between n_a and n_b), in an individual n -manifold with unresolved substates, or in an individual (Stark) substate of a given n -manifold. Fig. 1b shows the most recent, inclined laser beam-particle beam geometry we have been using for double-resonance CO_2 laser production of H(high n) and He(high n) atoms. Unlike our previous collinear-beam geometry^{2,12} the inclined geometry allows us to have each laser beam interact with the atoms in only one section of the beam apparatus. Fig. 1a shows a microwave cavity being used to ionize the Rydberg atoms for subsequent detection of the resultant fast ions. We also use ionization in a static axial electric field for detection (not shown in Fig. 1).

SCHEMATIC OF APPARATUS (SIDE VIEW)

Fig. 1b - A side view of a part of the apparatus shown in Fig. 1a. It shows the geometry of the two continuous CO₂ laser beams which cross the fast atomic beam in electric field regions F₁ and F₃, respectively.

Fig. 2 - Upper trace: A "single-resonance" Stark-tuned spectrum displaying CO₂ laser-driven transitions in F₁ originating on a large number of H(n=10) substates. Lower trace: A "double-resonance" spectrum displaying only those transitions out of the H(n=10) Stark substate (10,0,9,0).

Fig. 2 shows two Stark-tuned spectra² obtained with one collinear CO₂ laser beam driving transitions between substates of two different pairs of n-manifolds: n=7 and n=10 in F₁ and n=10 and n=33,34 in F₃. Each n-manifold has $n(n+1)/2$ Stark substates. The "single-resonance" spectrum (detected with one lockin amplifier) registered the many $n=10 \rightarrow n=33,34$ transitions in F₃ that were connected by appreciable matrix elements. The "double-resonance" spectrum (detected simultaneously with a second lockin amplifier) registered only those driven in F₃ out of the single H(n=10) substate (10,0,9,0). It was previously "tagged" by the laser-

driven transition $(7,0,6,0) \rightarrow (10,0,9,0)$ in F_1 . Obviously, the double-resonance method allows isolation of individual substates.

Fig. 3- Dotted lines: Experimental $\Gamma_I(F)$ curves for the hydrogen Stark substates 1: $\{40,39,0,0\}$; 2: $\{40,38,0,1\}$; 3: $\{40,38,1,0\}$; 4: $\{40,37,1,1\}$; 5: $\{40,37,2,0\}$; 6: $\{40,36,2,1\}$. The tic marks are discussed in Ref. 13. Solid lines: Theoretical curves calculated with use of a semiempirical, asymptotic formula (Eq. (6) of Ref. 14) with allowance for reduced-mass corrections. Squares: Numerical theoretical results calculated by Damburg and Kolosov for an infinitely massive nucleus.

These methods permitted the first precise measurement of the static electric field ionization rate $\Gamma_I(F)$ for resolved hydrogen Stark substates.¹³ Fig. 3 shows some experimental results (dotted lines) compared with numerical theoretical calculations (squares) and with calculations based on a semi-empirical analytic formula (solid lines)¹⁴. Since the six $n=40$ substates have large positive values of the "electric quantum number" $n_1-n_2=34-39$, they ionize at relatively large values of the scaled field, $n^4F \approx 0.2-0.3$ au. Because of the quantal tunneling, ionization at rates $\approx 10^7 \text{ s}^{-1}$ occurs at values of n^4F about 20% below the classical ionization thresholds calculated and displayed graphically by Banks and Leopold.¹⁵ This behavior was also observed¹³ for a state $(30,0,29,0)$ having a large negative value of $(n_1-n_2)=-29$. An important conclusion of this work is that quantal tunneling lowers the observable ionization threshold in static fields ($\omega=0$) as much as 20% below the classical threshold.

4. Quantal vs. Classical Theories of Multiphoton Ionization. - It is an interesting and important question to determine whether tunneling would significantly lower the ionization threshold in an oscillatory field, $\omega \neq 0$. We are explicitly investigating this question in current experiments. Theoretically, the answer to this question is unclear. Keldysh¹⁶ introduced a semiclassical model that attempts to treat non-perturbatively tunneling and multiphoton ionization as different limiting cases of the same physical process, namely ionization of an atomic electron by an intense electromagnetic field. He introduced the use of a parameter γ that is the ratio of

the time required for the electron to tunnel through the instantaneous barrier to the period of the field; γ much smaller and much larger than one correspond, respectively, to the adiabatic tunneling and multiphoton ionization limits. In atomic units, $\gamma = \omega/nF_0$ for hydrogen. Various assumptions made in his early work with this model were improved by Perelomov et al.,¹⁷ but it is not obvious that the model has any validity for the case of very high-order microwave ionization of H(high n) atoms. The large density of Rydberg states (proportional to n^3) means that a large number of multiphoton resonances between intermediate states are possible on the way to the continuum; the model does not handle these. Nevertheless, a prediction of this model in the adiabatic limit ($\gamma \ll 1$) is that ionization in the oscillatory field has a lower threshold than in a static field. As will be seen below, this is also a prediction of calculations based on classical theory, which ignores tunneling.

Other papers of this Colloquium by Brandi and by Mainfray deal with theoretical and experimental investigations, respectively, of the validity of the Keldysh model of ionization. Reiss¹⁸ has also reinvestigated it theoretically. It has not been specifically applied to the problem of microwave ionization of atoms.

Leopold, Percival, and Jones (Refs. 6-8, see also a review of their work in another paper of this Colloquium by Richards) developed an entirely classical theory of microwave ionization and excitation. The theory depends on two parameters: (1) ω/ω_{at} , the ratio of the frequency of the field to the atomic orbital frequency, and (2) n^4F_0 , the scaled peak electric field strength. In addition, they introduced the use of a "compensated energy" for the orbiting electron in the oscillatory field to decide whether ionization did or did not take place. From considerations of the magnitudes of classical actions compared to $\hbar$, they concluded that tunneling played a negligible role in the microwave experiments on $n=66$ atoms.^{19,20} Since static ionization experiments show us that tunneling is very important in the $\omega=0$ limit, however, it is unclear at what values of the scaled frequency ω/ω_{at} and the scaled field strength n^4F_0 tunneling ceases to become important.

5. Typical Parameters for Microwaves Used in the Experiments. - Microwave sources and plumbing span the range from 1 GHz to more than 500 GHz. Generally, as the frequency goes up, the available output power of sources goes down, all components get smaller, and cost generally increases. There are a large number of microwave frequency bands; operation in a new band usually means that new components must be acquired. For this reason, the microwave experiments reviewed in this paper have not yet been performed at a large number of widely separated frequencies. Most have been in the range 7-12 GHz, where sources and components were available.

Table 1

n	F_0 for $n^4F_0=0.1$	$E/\hbar\omega$	$\Delta E_1/\hbar\omega$	$P_{cav}(W)$	$P_{wg}(W)$
10	51.4 kV/cm	3292.1	658.0	10 kW	3.1 MW
20	3.2 kV/cm	823.0	82.3	40 W	12 kW
30	0.6 kV/cm	365.8	24.4	1.5 W	0.4 kW
40	0.2 kV/cm	205.8	10.3	0.2 W	0.05 kW
50	0.08 kV/cm	131.7	8.8	0.03 W	7 W
60	0.04 kV/cm	91.4	3.0	0.006 W	2 W
70	0.02 kV/cm	67.2	1.9	0.002 W	0.5 W
80	0.01 kV/cm	51.4	1.3	0.4 mW	0.1 W
90	8 V/cm	40.6	0.9	0.3 mW	0.07 W

Table 1 shows some useful numbers for 10 GHz microwave ionization experiments of H(high n) atoms. This frequency is near those used in most reported experiments. The columns show, respectively: (1) the principal quantum number, (2) the electric field strength F_0 needed to produce a scaled field strength $n^4F_0=0.1$ au, approximately the value observed empirically to produce appreciable ionization (notice that n^4F_0 is the classical field strength parameter mentioned above); (3) the ratio of the binding energy of the level to the 10 GHz photon energy, or the number of photons energetically needed to get to the zero-field ionization limit; (4) the

ratio of the energy separation between the levels n and $(n+1)$, or the number of 10 GHz photons needed to drive a $\Delta n=1$ transition (this ratio is also the inverse of the classical scaled frequency ω/ω_{at} mentioned above); (5) the approximate power that must be coupled into (and dissipated in) a typical TM_{020} -mode cylindrical cavity [unloaded $Q \sim 10^4$ and coupling voltage-standing-wave-ratio (VSWR) ~ 1] in order that the value of F_0 on the axis of the cavity equal that shown in column 2; (6) the approximate power that must be transmitted through a TE_{10} -mode X-band (8-12 GHz), rectangular waveguide in order that the value of F_0 in the waveguide equal that shown in column 2.

If we adopt ~ 10 W as an affordable upper limit on the power available from a tunable, continuous microwave source (such as the combination of a low-power oscillator and a traveling-wave-tube amplifier), waveguide ionization experiments are restricted to $n \geq 50$ and cavity experiments to $n \geq 25$. This means that inter- n transitions require on the order to ten photons, whereas ionization requires on the order of one hundred photons. Higher power, fixed frequency sources such as magnetrons are available in this and higher frequency ranges, but they would be best suited to experiments using resonant cavities. With them, one would be unable to study the frequency dependence of atom-field interactions. As we shall see, such studies are extremely important and have uncovered resonant behavior that is only partially understood.

6. Previous Ionization Experiments and Some New Results. - Bayfield and Koch¹⁹ first reported eight years ago the observation of microwave ionization of H(high n) atoms in the n -band $63 \leq n \leq 69$. Fig. 4 shows their data for the ionization probability at 9.91 GHz (solid circles), which corresponds to a scaled frequency $\omega/\omega_{at} \approx 0.43$. The atoms adiabatically entered and exited the microwave field inside a TM_{020} -mode cavity and were exposed to about 10^3 oscillations. The open circles show the classical, Monte-Carlo theoretical results of Leopold and Percival.^{6,7} Since, to date, these are the only calculations in quantitative agreement with the ionization experiments, the classical theory must be examined further as a viable model for the interaction of Rydberg atoms with intense microwaves. The horizontal axis is in units of the parameter γ , which in the classical theory is the ratio $(\omega/\omega_{at}) / (F_0/F_{at})$ of the classical frequency and field strength parameters. It is numerically equal to the Keldysh parameter, but its interpretation is very different because the classical theory obviously does not account for tunneling ionization.

It was found⁶⁻⁸ that the classical electron orbits in the intense field could be grouped into four different classes C1-C4. Each could be recognized by its distinctive spatial trajectory or by a plot of its "compensated energy" E_c vs. time. The latter are shown in Fig. 5. Motions C1 lie on an invariant torus in phase space and may never ionize; C2 ionize rapidly and directly into the continuum; C3 ionize more slowly and pass through at least one intermediate, extra-highly-excited (EHE) state; C4 pass through one or more EHE but do not ionize during the time scale of the calculation. Notice that C3 and C4 exhibit de-excitation (downward) transitions as well as excitation (upward) transitions.

The classical, non-linear dynamics of the ionization process is complicated. It is not clear how one would construct a quantum mechanical theory of microwave ionization that would reproduce this behavior and give physical insight into the different types of electronic motions. Stable and unstable (escaping) classical trajectories may co-exist for different initial conditions at the same values of the classical parameters, and both excitation and de-excitation transitions seem to be important. It appears from the plots in Fig. 5 that the electron may not end up with much kinetic energy after ionization, but this was not explicitly evaluated in the classical calculations.⁶⁻⁸ With use of a strong-field, quantal, perturbative theory, Gersten and Mittleman²¹ attempted to estimate how far into the continuum the electron would be ionized, but the validity of their theory for the microwave experiments was very uncertain. They tentatively concluded that, on average, twice the minimum number of photons would be absorbed. Since the Rydberg atoms in the experiments were bound by only tens of meV, their result, if true, means that the ionized electrons would have only this amount of kinetic energy. Recent laser multiphoton ionization experiments (reviewed in another talk of this Colloquium by Mainfray) have determined the relative probability for ionization of atoms by more than the minimum number of photons, but the theoretical interpretation of these

Fig. 4 - Closed circles: The measured probability (Ref. 19) for ionization of H(high n) atoms in the n -band $63 < n < 69$ by a 9.91 GHz microwave field as a function of the parameter γ , which is numerically equal to both the "Keldysh" parameter (Ref. 16) used in a quantal theory and to the ratio $(\omega/\omega_{at})/(F_0/F_{at})$ of the two parameters used in a classical theory (Refs. 6-8) of the interaction of hydrogen atoms with an oscillatory electric field. Open circles: Results of Monte-Carlo calculations (Refs. 6,7) that employed the classical theory to model the experiment of Ref. 19.(taken from Ref. 6)

experiments is still in progress.

It may be possible to use electron spectroscopy to gain information about the distribution of kinetic energy of electrons ionized by microwaves. Showing the feasibility of this type of measurement was a by-product of a very recent collaborative experiment at Argonne National Laboratory undertaken by our Yale group and a group at the Argonne Dynamitron Laboratory. More will be said at the end of this article about the experiment, whose data are still being analyzed.

7. Frequency-dependent Effects.- The resonant response of H(high n) atoms to certain ranges of microwave frequency was observed by Bayfield, Gardner, and Koch²⁰. In order to be able to sweep the frequency over the range ~ 9.6 -11.4 GHz, the atom beam was directed through an X-band, $TE_{1,0}$ mode waveguide. As was explained earlier via Table I, available microwave powers < 20 W meant that ionization of atoms laser-excited into individual n -levels could be studied for $n \leq 45$. Fig. 6 shows ionization curves for each level between $n=45$ and $n=57$. Fig. 7 shows, just for $n=48$, an ionization curve (lower frame) and an excitation curve (upper frame). The latter was taken in a "two microwave field" experiment that was sensitive only to atoms that were excited to $n > 48$ bound levels, but not to the continuum. Both curves in Fig. 7 were taken at a microwave power of 8 W, which corresponds to $F_0 \approx 80$ V/cm. Each curve

Fig. 5 - Plots of the "compensated energy" E_c vs. time for four different types of orbits C1-C4 obtained with Monte-Carlo calculations (Refs. 6-8) that were based on a classical theory of the interaction of hydrogen atoms with an oscillatory electric field. The calculations followed the electron during several hundred field oscillations. When $E_c < 0$, the electron is still bound to the proton. When $E_c > 0$, the electron is ionized. EHE stands for an extra-highly-excited Rydberg state with principal quantum number about a factor of five larger than the initial one. (taken from Ref. 6)

in Fig. 6 was recorded at a different microwave power empirically adjusted to limit the fraction of atoms ionized to 1-30%. The data in both figures lie in the following ranges of the classical parameters: (1) $0.13 < \omega/\omega_0 < 0.26$ (or in the near-adiabatic frequency regime), and (2) $F_0/F_{at} = n^4 F_0 \approx 0.09$ au, or significantly below the classical ionization thresholds $F_0/F_{at} = 0.13-0.38$ au^{8,15}.

The ~ 8 mm diameter beam-entrance and -exit holes perturbed the waveguide electric field distribution so that F_0 was within $\sim 5\%$ of its peak value only over a fraction, perhaps half or less, of the 1 cm separation between the opposing faces. This meant that the ~ 10 keV atoms experienced a transit-time-induced frequency spread of 0.05 GHz or more. The 0.5-1.0 GHz widths of the resonances in Figs. 6 and 7 are significantly larger than this, so they may be physically important. The absolute rate scales in Fig. 7 show that multiphoton ionization and excitation may proceed at comparable rates; which is dominant depends sensitively on the frequency.

These experimental results stimulated theoretical activity. Delone et al.¹¹ advanced a diffusion model to characterize the microwave ionization, but their

Fig. 6- The measured ionization of H(high n) atoms with each principal quantum number n between 45 and 57 as a function of the microwave frequency between 9.6 and 11.4 GHz. The vertical scales are relative units different for each n . The vertical bars are worst-case estimates of the fluctuations in the signals that were caused by frequency-dependent variations in the peak microwave electric field strength experienced by the atoms inside the $TE_{1,0}$ mode waveguide. The microwave power incident on the waveguide was kept approximately constant for each n -value but was readjusted with each change in n to keep the peak fractional ionization in the range 1-30%.

Fig. 7- A comparison of the frequency dependence and absolute rates for microwave multiphoton excitation (a) and ionization (b) of H($n=48$) atoms. The open circles in (a) are background signals. Single- and double-headed arrows show the position of multiphoton transition frequencies between a field-free $n=48$ state and field-free $n=49$ and $n=50$ states, respectively, for the number of photons shown above the arrows.

estimated diffusion time was criticized by Meerson et al.¹⁰, who proposed an ionization model based on the stochastic instability of a classical, non-linear oscillator. In pointing out that their own classical, Monte-Carlo trajectory results were directly related to the general problem of the onset of stochastic motions in non-linear dynamical systems, Percival et al.⁶⁻⁸ concluded that "much more work needs to be done...to obtain a clear picture of the physical process responsible for ionization." In a recent review article Zaslavsky amplifies and extends these points of view and "...show(s) the possibility of the existence of a stochastic mechanism for the ionization of atoms." (Ref. 9, p.236).

A possible, more fundamental problem concerns the nature of stochasticity in quantum systems. Again quoting Zaslavsky (Ref. 9, p.160):

At present, the number of papers considering stochasticity in quantum systems is rather large. Not only is there no understanding of the general situation but there is no more or less clear answer to even a single, the "simplest", problem, either. Among them are the fact that not even a single model has been accurately solved completely, that there are no exact results, that there is the necessity to obtain results true for long times, and that there are difficulties in using computers.

Zewail concludes, similarly, that the underlying theoretical is "...the question of what precise relationship, if any, exists between classical chaos and quantum mechanical motion. Clearly we still do not know the precise meaning of quantum ergodicity." (Ref. 22, p.33).

Since the classical and quantum mechanical motions of the field-free hydrogen atom are known exactly, the hydrogen atom in an electromagnetic field may be a useful testing ground for intense-field studies of these issues. One of them is obviously what influence quantal tunneling may have on the onset of chaotic and escaping motions in non-linear systems.

8. New Hydrogen and Helium Experimental Results.- At Yale, our group has recently completed running some new experiments on microwave ionization of hydrogen and helium Rydberg atoms. The goals were rather modest: We were interested in comparing the F_0 -dependence of the ionization of a simple non-hydrogenic atom with that of hydrogen. The apparatus was similar to that shown in Figs. 1a,b, and a 9.91 GHz TM₀₂₀-mode cavity was used to produce the ionizing microwave field. Analysis of these new experiments is still in progress, and the author appreciates the willingness of his colleagues at Yale, D. Mariani and W. van de Water to have some of our preliminary results and conclusions presented here.

For a variety of reasons, we chose to work with n^3S_1 Rydberg states of helium. We shall compare results for atoms in different n -manifolds by using the scaled electric field n^4F , where $n^* = [n - \delta(L, S)]$, the effective quantum number, equals the difference of the principal quantum number n and the quantum defect $\delta(L, S)$, which depends mainly on the orbital L and spin S angular momenta of the particular level. Helium n^3S_1 Rydberg levels have the largest quantum defect, $\delta(0, 1) = 0.297$. Since it will be useful to compare their microwave ionization behavior to that in a static field, we show in Fig. 8 a static field ionization curve for He(41 3S_1) atoms prepared by double-resonance, CO₂ laser techniques^{2,12,13} in fields F_1 - F_5 in Figs. 1a,b. Fig. 8 shows the fraction of an ~ 12 keV beam of these atoms that survived passage through the 8.3 cm-long ionizing field F_5 in Fig. 1a. It was recorded using the same methods previously used for precise measurement of field ionization rates of hydrogen Stark substates¹³. We notice a sharp ionization threshold (drop in the curve) at $n^4F \approx 1/16$ au, which is the "saddle-point" ionization threshold that characterizes ionization of non-hydrogenic, low- $|m|$ Rydberg states that follow adiabatic paths to the continuum²³.

Fig. 9 shows microwave ionization curves for H($n=41$) atoms (with an unmeasured distribution of substates) and He(41 3S_1) atoms. Each curve shows a signal proportional to the fraction of atoms ionized during their passage through the cavity;

Fig. 8.- The measured fraction of He($4l\ ^3S_1$) atoms in a 12 keV beam that survive passage through the 8.3 cm-long ionizing field F_5 shown in Fig. 1a.

Fig. 9 - Microwave ionization curves for H($n=4l$) atoms (dashed curve) and He($4l\ ^3S_1$) atoms (solid curve). The horizontal scale is proportional to the microwave power incident on the 9.91 GHz, TM_{020} -mode cavity; therefore, the scale of the peak amplitude of the microwave electric field F_0 is non-linear. The absolute F_0 -scale is uncertain by about 10%. The H and He atoms were exposed to about 6.5×10^2 and 1.3×10^3 cycles of the microwave field, respectively. The signals saturate for $n \cdot 4 F_0 \gtrsim 0.12-0.14$ au.

the two curves have been approximately normalized. For $n=41$, the 9.91 GHz frequency corresponds to a frequency ratio $\omega/\omega_{at} \approx 0.1$, or in the "near-adiabatic" regime. Even though the difference in zero-field binding energies of the atoms is very small, $\Delta E \approx \delta(0,1)/n^3 \text{ au} \approx 0.95 \text{ cm}^{-1} = 28 \text{ GHz}$, or fractionally only 1.4%, the two ionization curves are very different. The hydrogen ionization threshold is approximately $n^4 F_0 = 0.08 \text{ au}$, and there appears to be some structure in the curve near threshold. The ionization signal is saturated (rate $> 10^8 \text{ s}^{-1}$) at $n^4 F_0 \approx 0.13 \text{ au}$. For helium, however, the ionization signal rises at much smaller values of $n^4 F_0$ and is not saturated until a somewhat higher value than that for hydrogen. Fig. 10 shows another hydrogen curve, this time for $n=27$. For this n -level, the frequency ratio is $\omega/\omega_{at} \approx 0.03$, or even more into the adiabatic regime. In terms of the scaled field $n^4 F_0$, the $H(n=27)$ curve is very similar to the $H(n=41)$ curve, except for the lack of possible structure near threshold.

Fig. 10 - A microwave ionization curve for $H(n=27)$ atoms that was taken under the conditions described in the caption to Fig. 9. The signal saturates for $n^4 F_0 \gtrsim 0.12 \text{ au}$.

Let us analyze the hydrogen curves first in terms of the classical, Monte Carlo theoretical results⁸. Fig. 11 shows, for a number of values of the scaled field $F_0/F_{at} = n^4 F_0$, a composite of their calculations of the percentage of ionization vs. the frequency ratio ω/ω_{at} . (We point out that more extensive calculations at fixed $F_0/F_{at} = 0.08$ and on a finer mesh of frequency ratios ω/ω_{at} between 0.3 and 1.4 shows several resonances in the ionization probability (Ref. 8, Fig. 3) not unlike those observed experimentally--see Figs. 6,7 above. Unfortunately this range of ω/ω_{at} is slightly above the range 0.13-0.26 of the experiments²⁰.) We notice that for $\omega/\omega_{at} < 0.1$ (or to the left of $\lg(\omega/\omega_{at}) = -1$ on each horizontal scale), they predict that negligible ionization takes place until $F_0/F_{at} > 0.1$. Recall that the measured hydrogen curves in Figs. 9, 10 show the onset of ionization signals at $F_0/F_{at} \approx 0.08$. A preliminary analysis leads to about a 10% uncertainty in our determination of the absolute field strength F_0 in the cavity in terms of the measured power incident on it. Therefore, we have some preliminary evidence that the observed onset of ionization occurs at somewhat lower values of F_0/F_{at} than those predicted by the classical theory⁶⁻⁸. It is highly tempting to view this as evidence for the importance of tunneling, but we shall avoid this categorical assertion until we have made a complete analysis of our data and until a larger

Fig. 11 - A composite of the results of classical Monte-Carlo calculations (Ref. 8) at a number of values of the scaled field F_0/F_{at} of the percentage ionization of hydrogen atoms in an oscillatory electric field whose frequency ω is related to the atomic orbital frequency through the ratio ω/ω_{at} . Notice that the ionization threshold for a nearly static field is near $F_0/F_{at} = 0.13$ au and that the ionization threshold for oscillatory fields with $\omega/\omega_{at} \approx 1$ is about a factor of two lower. The classical calculations ignore the effects of quantal tunneling, which lowers the observed static ionization threshold (Ref. 13) by 10-20% depending on the particular Stark substate studied. (taken from Ref. 8)

Fig. 12- A microwave ionization curve for $\text{He}(28\ ^3S_1)$ atoms that was taken under the conditions described in the caption to Fig. 9. The signal saturates for $n*4F_0 > 0.16$ au.

number of classical calculations are available in the threshold region.

Now we move to a discussion of the helium microwave ionization data. Fig. 12 shows an ionization curve for He($28\ ^3S_1$) atoms. Similar to the He($41\ ^3S_1$) curve shown in Fig. 9, the ionization signal rises at very low values of n^4F_0 , but the He($28\ ^3S_1$) curve has a plateau (and even a slight dip) between about $n^4F_0 = 0.05$ - 0.1 au, rises again above this value, and does not saturate until about 0.16 au. Fig. 13 shows an ionization curve for He($28\ ^3S_1$) over a much smaller

Fig. 13 - A microwave ionization curve for He($28\ ^3S_1$) atoms that emphasizes the threshold, low- F_0 region of Fig. 12. The origin of the "bump" is explained in the text. The signal for $n^4F_0 > 0.02$ au is not saturated; it is a plateau unobservable on the scale of Fig. 6.

range of n^4F_0 -values near the onset of ionization. Notice that F_0 scales of the two figures are so different that the very interesting threshold structure observable in Fig. 13 is compressed and unobservable on the scale of Fig. 12. We see that the ionization signal in Fig. 13 begins very sharply at $n^4F_0 \approx 0.011$ au, or $F_0 \approx 100$ V/cm, rises to an local maximum, decreases, rises again more slowly, and then reaches a local plateau.

We are able to explain the "bump" near 100 V/cm in terms of known features of the helium spectrum. Fig. 14 shows a "Stark map" of $m=0$ triplet Rydberg levels. The calculation was performed on an (n, ℓ, m_ℓ) basis by Dr. van de Water of our group using matrix-diagonalization programs adapted from those kindly supplied by Dr. Zimmerman²⁴. The magnetic fine-structure of helium is negligible on the scale of the map and was ignored in the calculation. Notice that the adiabatic continuation of the He($28\ ^3S_1$) level undergoes its first avoided crossing with the uppermost $m=0$ triplet level (which is the adiabatic continuation of the $(n=27, \ell=26, m_\ell=0)$ level) rising from the $n=27$ manifold near 110 V/cm, which is within estimated experimental error of the position of the threshold "bump" in the ionization curve. Since the frequency ratio $\omega/\omega_{\text{at}} \approx 0.03$, the oscillation of the applied electric field is adiabatic with respect to the motion of the electron. The "Stark map" in this case is analogous to the molecular potential curves that one obtains in a Born-Oppenheimer separation of the slow internuclear motions from the fast electronic motions in a molecule. In a molecule, or in a quasi-molecular treatment of an atom-atom collision, the internuclear motion produces dynamic terms which couple states belonging to different adiabatic molecular potential curves. In the present case, the oscillatory field moves the system back and forth along the Stark potential curves and may cause the system to jump (make diabatic, or Landau-Zener transitions²⁵ from one potential curve to another.

Fig. 14- A preliminary "Stark map" for some $m=0$ triplet Rydberg levels of helium that was calculated by Dr. W. van de Water at Yale with use of matrix-diagonalization programs adapted from those kindly supplied to us by Dr. M.L. Zimmerman (Ref. 24). It was computed on an (n, ℓ, m_ℓ) basis of 106 states in the $n=26-29$ manifolds. Magnetic fine-structure is unobservable on the scale of the figure and was neglected.

It appears, therefore, that partially diabatic transitions at this first and at other avoided crossings transfer population from the initial level onto other Stark substates. The remarkable thing is that the system is then able to proceed all the way to the continuum at this rather low field strength. Since the atom experienced approximately 10^3 oscillations of the field, however, it had many opportunities to make such transitions. We note that the first avoided crossing between adiabatic levels in neighboring manifolds, n and $(n-1)$, occur at an F_0 -value that scales as n^{-5} .

In additional experiments with $n=26, 29$ $\text{He}(n\ 3S_1)$ atoms, we observed threshold "bumps" and "plateaus" similar to those shown for $n=28$ in Figs. 6, 7. For these other n -values, which also have adiabatic frequency ratios $\omega/\omega_{\text{at}}$, the bumps show up at F_0 -values corresponding to the first avoided crossing. We did not observe a threshold "bump" with $\text{He}(41\ 3S_1)$ atoms, however. It is tempting to explain this behavior by the less adiabatic frequency ratio $\omega/\omega_{\text{at}} \approx 0.1$ for $n = 41$. If this is so, one would have an effect analogous to the breakdown of the Born-Oppenheimer adiabatic approximation in molecules.

Though our preliminary analysis explains, at least partially, the origin of some threshold bumps, we cannot explain quantitatively the location of plateau regions over which some ionization curves are locally independent of the value of F_0 . Our experiments on microwave ionization of non-hydrogenic atoms have displayed fascinating behavior that is very different from that observed in hydrogen atoms having nearly the same initial binding energy. One may speculate that the hydrogen-like, high- $|m|$ states of a Rydberg atom like helium may display microwave ionization behavior more like that of hydrogen. Since the electric field interaction conserves $|m_\ell|$ (at least in the limit of negligible magnetic fine-structure),

the oscillatory electric field would not induce transitions between different $|m_l|$ -states. Stray fields or collisions could do this, however.

We offer an important conclusion: Nearly all the theories treating microwave ionization have been based on a non-relativistic treatment of hydrogen. Since we have observed dramatic differences between hydrogen and non-hydrogenic Stark states, it is inappropriate to use non-hydrogenic Rydberg atoms in experiments designed to test theories based on hydrogen. Nevertheless, the non-hydrogenic effects will be interesting to study in detail in their own right, but one will need to develop additional theory.

9. Structure of H(high n) Atoms in an Intense Microwave Field.- Thus far, we have emphasized questions of atomic transitions driven by intense microwaves, from one bound state to another or from a bound state to the continuum. In this section, we consider the problem of atomic structure, or what are the (quasi-)bound energy levels of the hydrogen atom in the intense microwave field. Blochinzew²⁶ first investigated the quantal structure of the Hamiltonian Eq. (2). More recent treatments of the "AC-Stark effect" have included those of Autler and Townes²⁷, of Bakshi et al.²⁸, of Hicks²⁹ and Hicks et al.³⁰, and of a number of other theorists whose work has been reviewed recently by Delone et al.³¹ and by Bayfield^{4,5}. For the degenerate states of the hydrogen atom, these theories predict that one effect of an intense, linearly polarized oscillatory field is to produce quasi-energy satellites (also called dressed states, photon replicas, etc.) displaced by integral numbers of photons $\pm p\hbar\omega$ above and below the zero field level. The amplitude of the satellite of order p is proportional to

$$J_p(dF_0/\hbar\omega), \quad (3)$$

the value of the Bessel function of order p whose argument is the ratio of the peak dipole energy dF_0 to the photon energy $\hbar\omega$. As in the static Stark effect, the permanent dipole moment d is created from opposite-parity atomic states that are mixed by the field. The Bessel function weighting of the satellites is a general feature of intense-field problems: they appear, for example, in spin systems in intense, oscillatory magnetic fields³² and in electron-atom collisions in the presence of an intense laser field³³.

Twenty years ago, Baranger and Mozer³⁴ proposed using spectral-line-satellites produced by the AC-Stark effect as a diagnostic probe of high-frequency electromagnetic oscillations in a plasma. This motivated a significant amount of experimental and theoretical work (for useful lists of references see Refs. 35 and 36). The Ph.D. thesis of Hicks²⁹ includes the report of a nice optical spectroscopy experiment on a hydrogen plasma, but to this author's knowledge, it is available only as a laboratory report. He measured the Balmer spectrum emitted by the hydrogen plasma inside a Geissler tube that was collinear with the microwave electric field inside a rectangular, TM_{010} -mode cavity. He was able partially to resolve the microwave-induced satellites on each side of the $H_\alpha, H_\beta, H_\gamma, H_\delta$ lines and to obtain excellent results with a theory based on the work of Blochinzew.

The agreement between experiment and Blochinzew-type theory has generally been much less impressive for experiments which used internal fields inside a plasma, particularly a turbulent one, to produce the satellites. An externally applied field was used in the published helium experiment of Hicks et al.³⁰ and in the hydrogen experiment just mentioned²⁹. Nee and Griem³⁵ observed internal-plasma-field-induced $p=\pm 1$ satellites of $n=13 \rightarrow 12$ and $n=12 \rightarrow 11$ infrared transitions in hydrogen but found that the absence of higher-order satellites could not be explained by a Blochinzew-type theory based on a single-frequency, constant amplitude electric field $F(t) = F_0 \cos \omega t$. They developed a theory to model the multimode fields of a turbulent plasma and obtained reasonable agreement with their experimental results. We note the experimental conditions described in Eq. (4.7) and Table I of Ref. 35 corresponded for $n=13$ to $F_0/F_{at} \approx 0.03$ au and $\omega/\omega_{at} \approx 0.05$. These values are not too far below those needed for ionization processes to become important. Their theory³⁵

ignored such processes. Sanchez and Bengtson³⁷ observed Stark-broadening and satellites of lines in a turbulent helium plasma and found existing theory incapable of explaining their results quantitatively.

10. Fast-beam Studies of Hydrogen Spectra in Intense Microwaves.— Bayfield³⁸ first proposed a fast-beam, laser experiment for production and detection of satellite levels of H(high n) atoms in a single microwave field. The present author³⁹ demonstrated the feasibility of this class of experiments with an important modification: The fast-beam was passed through two spatially-separated regions of microwave field whose strengths could be adjusted independently. CO_2 laser photons were used to drive transitions from initial $n_i=10$ levels with an unmeasured distribution of sub-states to final high n_f levels in the presence of the first microwave field, which produced the satellite structure being investigated. The second field was used to detect by microwave ionization those atoms that were pumped by both the laser and microwave photons in the first field. Thus, the satellite-producing field could be weaker than that needed to ionize the atoms. The frequency of the continuous CO_2 laser was Doppler-tuned in the atomic rest frame by varying the beam velocity. Since transitions to $n_f > 30$ levels were studied, we expect that if the effective dipole moment d of the atoms in the first field scaled as n^2 , the moment $d(n_f)$ of the final level was at least an order of magnitude larger than the moment $d(n_i=10)$ of the initial level. Therefore, the ratios of $dF_0/\hbar\omega$ that enter into the Bessel functions in Eq. (3) were much larger for the final level, and its satellites played a more important role than those of the initial level.

The results of that first unpublished experiment³⁹ were included in a recent review², from which Fig. 14 is taken. The hatched curves show experimental data for the production of the $p_f = +1, +2$ satellites of H($n_f=48$) in a 9.91 GHz field with peak amplitude $F_0(A)$. The abscissa of the figure is a quadratic $F_0(A)$ scale (proportional to the power dissipated in the microwave cavity) normalized to the field $F_0(\text{ion}) \approx 90$ V/cm that was observed to ionize the H($n_f=48$) atoms. We see that the $p_f = 1, +2$ signals peak at values of $F_0(A)$ less than 10% of $F_0(\text{ion})$. The dashed curves show theoretical signal curves obtained using the form of the amplitude given by Eq. (3). The $p_f = +1$ dashed curve was normalized by adjusting the magnitude and argument (magnitude of d_f) of the first maximum of J_1^2 to agree with the position and height of the peak of the experimental curve. The same normalization parameters were used with the function J_2^2 to plot the dashed $p_f = +2$ curve. The fitted dipole moment is $d_f = 2.6 \times 10^3$ au, or scaled down by n^2 , $d_f^2/n_f^2 \approx 1$ for $n_f = 48$. This is a rather reasonable result; the permanent dipole moment in the microwave field was about one scaled atomic unit (au/ n^2). According to Delone et al.³¹, the linear interaction of the atoms with the oscillatory field dominates the quadratic interaction when $n^4 F_0 \ll 0.06$ au, or $F_0 \ll 60$ V/cm for $n_f = 48$. Since in Fig. 14 $F_0(A) \lesssim 10$ V/cm, the experiment fulfilled this condition.

Bayfield et al.⁴⁰ recently published the results of a more extensive study of the spectra of H($n_i=44-67$) atoms in a 5.9-8.0 GHz microwave field. They used a "two-field" technique similar to the one mentioned above. Fig. 15 shows two of their spectra for excitation of $n_i=10$ atoms to satellites of $n_f=43-45$. The higher-power spectrum in the lower frame exhibits more complicated, overlapping series of satellites (up to $|p_f|=15$) compared to the lower-power spectrum in the upper frame. Since the zero-field hydrogen levels ($p_f=0$ satellites) are unequally spaced (n to $(n+\Delta n)$ spacing equal to $\Delta n/n^3$ au) and the satellites of a given n -level are equally spaced, the satellite patterns of different n -levels are out of step. In the limit of a huge number of satellites being appreciably populated in a strong field, one would expect the empty spaces between the zero-field levels to fill up with a kind of "lumpy continuum." The analogy²⁰ between the problems of multiphoton excitation and ionization of hydrogen atoms by a strong microwave field and of multiphoton dissociation of polyatomic molecules by an intense, infrared laser field should be mentioned here. The lumpy continuum in the present hydrogen problem is similar to the quasi-continuum of upper levels in the molecular problem (see Ref. 22 for a useful list of references).

Fig. 15.- Cross-hatched areas: Experimental signals for CO_2 laser-induced transitions from the $p_i=0$ satellite level of $\text{H}(n_i=10)$ to the $p_f=+1, +2$ satellite levels, respectively, of $\text{H}(n_f=48)$ in a 9.91 GHz electric field whose peak field strength was $F_0(A)$. $F_0(\text{ion}) \approx 90$ V/cm was the field strength observed to ionize the $\text{H}(n_f=48)$ atoms. Dashed curves: Theoretical curves based on Eq. (3) that are proportional to the square of the Bessel functions of order p_f . The normalization procedure is discussed in the text.

Bayfield et al.⁴⁰ measured excitation curves similar to those shown in Fig. 14, and saw no oscillations past the first maximum that one would expect according to the extrema and zeroes of Bessel functions, Eq. (3). They ascribed the deviation between the "theoretical" and experimental curves at higher values of F_0 as due to the n_f -satellite levels having a spread in the value of the dipole moment d_f . This would cause a spread in the value of the argument of each Bessel function amplitude, Eq. (3), and for each different d_f -value, the squared Bessel function curve would peak at a different value of F_0 . Some kind of average d_f -value can adequately explain the low- F_0 part up to the first maximum of each curve, but the spread in d_f -values changes the shape of each curve at higher F_0 -values. For n_f -levels between 44 and 67, and for frequencies 5.9–8.0 GHz, they found that the power at which each $p_f=-1$ satellite peaked was proportional to $\omega^{(3.0 \pm 0.5)} n^{(-4.0 \pm 0.2)}$, the corresponding value of d_f varies as $n^2 \omega^{-1/2}$.

Stehle used a quantal forward scattering method to model theoretically the two-frequency (laser + microwaves) excitation of $\text{H}(n_i)$ atoms to n_f -states. (The present author appreciates receipt of a pre-print on this work from Professor Stehle before its publication.) His theory makes a prediction that could be tested in future experiments: The excitation amplitude for each satellite is still weighted by its p th-order Bessel function, Eq. (3), but all satellite-amplitudes share a common factor $J_0(2dF_0/\hbar\omega)$. This new factor would make all satellites disappear together at values of F_0 corresponding to the zeroes of J_0 . If the experimental situation includes a spread of d_f -values, these would have to be taken into account in applying the theory. We mention that modification and even vanishing of the Lande g -factor in atomic magnetic resonance experiments in a strong oscillating magnetic field B_1 was observed and explained theoretically using the "dressed-atom" formalism

Fig. 16.- Excitation spectra (Ref. 40) in hydrogen atoms for CO_2 laser-driven transitions from satellite levels of the $n_i=10$ manifold to those of the $n_f=43, 44$, or 45 manifolds. Assuming that only the $p_i=0$ satellites of the $n_i=10$ manifold contribute, the additional numbers near the peaks label the orders p_f of the satellites populated in the n_f -manifolds. The fixed-frequency CO_2 laser radiation was Doppler-tuned in the atomic rest frame with variation of the kinetic energy of the atomic beam. Since lower energy atoms spent a longer time in the waveguide field region, the lower energy resonance peaks are more intense. The $p_f=0$ resonances contain large contributions from atoms pumped outside the waveguide field region. (taken from Ref. 40).

by Cohen-Tannoudji and Haroche many years ago (Ref. 32, Section II.B.) The effective g-factor (or, equivalently, the effective magnetic moment) in the intense field also displayed a zeroth-order Bessel function dependence on B_1 .

It is clear that future experiments will be much improved if one were to use $H(n_i=10)$ atoms prepared with a single value of d_1 , that is, in a single Stark sub-state. These methods have been developed for static field studies^{1,12,13} and must now be applied to the microwave studies. It is not yet known, however, what happens to a hydrogen atom prepared in a single, static Stark state when it passes into a microwave electric field. If the microwave electric field is collinear with the static field direction, the $|m|$ -quantum number will certainly be conserved, but it is not clear that the value of the static dipole moment will be. Perhaps a cleaner situation will be to superimpose collinear static and microwave electric fields, but some of the same uncertainties about whether the value of the dipole moment remains constant will still have to be addressed.

11. Mixed-field Studies.- If the hydrogen atom were exposed to a microwave field plus additional static or oscillatory, magnetic or electric fields, the problem would obviously be made more complex. Such a situation is beyond the scope of the present paper, but a few experimental and theoretical papers have been published which point the way to future studies. We have already mentioned above the plasma physics experiments which investigated modifications of field-free atomic spectra by internal, plasma fields. Some of these fields can be described as quasi-static and others as dynamic. Gallagher and Levine⁴¹ reported observation of satellites of H_β -lines in a turbulent hydrogen plasma that were ascribed to resonance effects between ortho-

gonal components of dynamic and quasi-static internal electric fields. These resonance effects, which produce spectra dramatically different from the Stark spectrum of each field acting independently, were first investigated numerically by Cohn et al.⁴² More recently, Gavrilenko and Oks⁴³ have analytically confirmed and extended the theoretical work of Cohn et al.⁴² They find that fields with fixed and random phases, respectively, exert different resonant action on the hydrogen spectral lines that start from levels with $n > 3$.

An important series of future experiments will use collision-free beams of hydrogen atoms laser-excited into individual (resonant-)satellite states produced by mixed, applied fields. This will be a cleaner experimental situation for studying the field-effects themselves than is provided by the internal fields in a turbulent plasma.

12. Additional Fast-beam Experiments.— Our group at Yale (Mariani, van de Water, and Koch) recently joined with a group (Gammel, Kanter, Schneider, et al.) at the Dynamitron Accelerator at Argonne National Laboratory to investigate further the influence of Rydberg atoms on measured energy spectra of "convoy electrons" produced by fast ions incident on a thin foil (for an extensive review of convoy electron studies, see Ref. 44). These electrons (called "cusp electrons" in ion-atom collisions in a gas target) emerge from the collision with nearly the same velocity and direction as the unscattered ions or forward-scattered ions that have captured target electron(s) into a bound state(s). For example, convoy electrons produced in 0.5 MeV H^+ -foil interactions emerge with a kinetic energy about a factor (m_e/m_p) smaller, or near 272 eV. The Argonne group showed previously⁴⁵ that electrons captured into bound Rydberg states could be ionized by electric fields in the electron spectrometer and elsewhere in the apparatus in such a way as to contribute to the apparent convoy-electron peak.

Since the analysis of the experiments is still in progress, we do not go into any detail here. We used a 9.91 GHz microwave cavity to ionize Rydberg atoms after their point of production in a thin (2-5 micrograms/cm²) carbon foil and before their entrance into the electron spectrometer used to observe the convoy-electron spectra. Because of its high frequency, the microwave electric field negligibly affected the "true" convoy electrons. The foil could also be electrically biased to shift the energy of the convoy electrons relative to those produced by microwave ionization of the Rydberg atoms. We were able to resolve clearly these two sources of electrons in collisions of 0.5-2 MeV H^+ and He^+ atomic ions and H_2^+ molecular ions with the foil. We are interested to see if an analysis of the data will give us preliminary experimental information on the continuum electron energy distribution produced in microwave ionization of hydrogen and helium Rydberg atoms.

13. Conclusions.— It is useful to summarize here the conclusions presented throughout the paper. The interactions of Rydberg atoms with an intense microwave electric field are not yet well understood. At the same field strength, multiphoton excitation and ionization can proceed at comparable rates, and each displays a different pattern of frequency-dependent resonances in hydrogen. To date, classical theory has furnished the only limited, quantitative agreement with hydrogen microwave ionization experiments. It also gives insight into some of the complicated motions of the atomic electron in the oscillatory field, but it ignores quantal tunneling processes that may be important in some cases. Tunneling has been shown to be important in static field ionization. There may be fundamental questions about a "stochastic mechanism" for ionization, but this has only been examined in a classical context which treats the hydrogen atom in the intense field as a non-linear, classical dynamical system. If quantal effects are important, new questions are raised because the nature of the onset of stochasticity in quantum systems is poorly understood. Because of their very different behavior in an intense microwave field, non-hydrogenic Rydberg atoms should not be used to test hydrogen theory. Though we are able to explain at least some threshold features of the microwave ionization of helium Rydberg atoms, we are not yet able to explain most of the structure in the ionization curve.

Quantum mechanics deals well with some aspects of atomic structure in the oscillatory field, but it has not yet been investigated carefully for fields strong enough to cause appreciable ionization. Recent theory predicts effects such as the disappearance of the satellite spectrum associated with a given n -level at certain values of the field strength F_0 . This could be interpreted as the field-induced modification and vanishing of the atomic electric dipole moment at certain values of F_0 , and, therefore, the electric resonance analog of the modification and vanishing of the Lande g -factor in magnetic resonance of atoms in a strong oscillating magnetic field.

Future microwave experiments will use available techniques for production of hydrogen atoms in individual Rydberg substates. These will allow more detailed studies of the atomic structure in the intense field as more refined excitation and ionization experiments.

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