Diffusionlike Aspects of Multiphoton Absorption in Electrically Polarized Highly Excited Hydrogen Atoms

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We have observed microwave transitions of overall order up to 16 in hydrogen atoms initially in $n=60$ lowest-energy Stark states. Increases in n as large as 4 occur with little change in the electric quantum number n_e. Thus hydrogen atoms electrically polarized along the direction of a strong linearly polarized microwave electric field respond one-dimensionally to this field by maintaining their polarization. At some microwave frequencies the final-state n distribution is smooth above threshold field values estimated for stochastic transitions.

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In the last few years the study of multiphoton transitions in highly excited atoms has been stimulated by the possibility of diffusive photon absorption.[1] Classical calculations for the driven one-dimensional hydrogen atom predict chaotic motion of the electron at high oscillatory driving forces, opening up the question of possible stochastic quantum statistics in real hydrogen atoms exposed to intense oscillating electric fields. The diffusive outward appearances of stochastic-driven bound-electron motion would include smooth atomic quasienergy spectra observed while the driving field is acting, and, although not as conclusive, smooth final-state quantum-number distributions observed after the driving field is switched off.

A special situation exists when the electron initially is in an extreme Stark state, far on one side of the atom's nucleus, and is in a linearly polarized external microwave field $F_\mu \cos\omega t$ directed along the atom's electric dipole moment. We discuss the first experiments ever carried out for this case. We find that the response of the driven polarized atoms is in essence one dimensional, and that the final-state distributions can appear diffusive. Earlier work has been with unpolarized atoms and unresolved final states.[2,3]

We used an optical double-resonance state-selection concept[4] to produce fast $n=60$ hydrogen atoms in well-defined Stark states with $n_e=n_1-n_2=-59$. A kiloelectronvolt-energy beam containing $n=7$ atoms produced by charge-exchange collisions in argon gas was first passed through a strong static electric field region where a collinear CO_2 laser beam selectively excited $(n,n_e,m)=(7,-6,0)$ atoms to the $(10,-9,0)$ state. The beam then passed into a weak static electric field region for further resonant excitation to the $(60,-59,0)$ state by the same laser. The data for this paper were taken at an atom kinetic energy of 19.00 ± 0.05 keV, a laser wavelength of 9.3294 μm, a strong-field strength of $23\,530\pm70$ V/cm, and a weak-field strength F_s of 5.50 ± 0.01 V/cm. The spectroscopic resolution for the laser transitions was 0.2 GHz. The atoms passed in 0.37 μs through a long-waveguide microwave uniform field region.[3] The atoms then passed through a field-ionization quantum-number analyzer and into a microwave cavity ionizer for detection of the produced fast protons.[2,3]

The static field F_s was present during the entire lifetime of each atom in the beam. A field strength set between 5 and 10 V/cm maintained well-defined values for n_e both before and after the second laser transition. This uniform field was produced motionally, the atom beam traveling in a magnetic field of 2.9 G produced by large coils. The Zeeman interaction with the atomic electron's spin was too small to be noticed.

Once all electric fields in the apparatus (including the microwave field) were oriented in the same direction to within 1°, we found that even at high power levels the microwaves did not induce the sizable n_e mixing characteristic of a reorientation of the $n=60$ atom dipole moment. The microwave-induced n-changing transitions involved the quantum-number linkage $n_e=1-n$. The evidence for this is the sharpness and location of the steps in the quantum-number analyzer scans of atoms exposed to the microwaves, as shown in Fig. 1. A 3% spreading out of the value of n_e would have noticeably broadened the steps in the direction of higher analyzer fields. This evidence of one-dimensionality in the atom's response to the microwave field connects experiment with one-dimensional quantum theoretical searches for stochastic behavior.

The frequency dependence of n-changing transitions at low microwave-power levels is shown in Fig. 2. The fact that these resonances appear where expected further establishes that the quantum-number analyzer measured principal quantum number. At a microwave-power level of 0.2 W ($F_\mu=8$ V/cm), the only up n changing occurring is resonant from $n=60$ to $n=61$, along with a weak tail extending down to 6.7 GHz. The origin of the tail may be transition-element terms of second and higher order in off-diagonal matrix elements,[6] but this remains to be established. At 0.5 W ($F_\mu=12$ V/cm), the $n=60$ to $n=61$ resonance broadens to a half-width of 100 MHz and has a peak value of 40% transition probability. Its tail then extends up to 8.0 GHz and typically contains 10% transi-

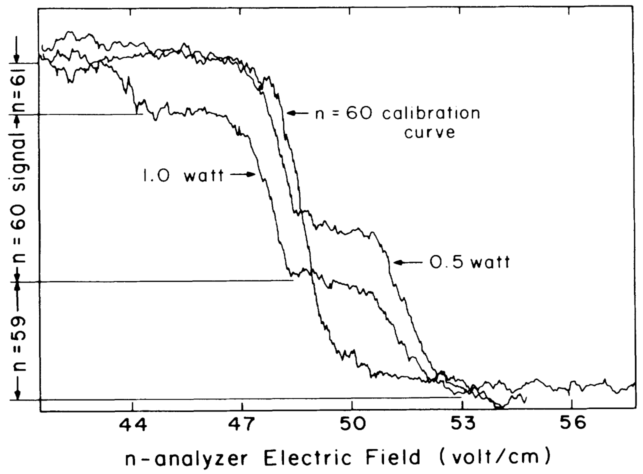

FIG. 1. Quantum-number analyzer scans of the n distributions produced at two microwave-power levels and for the microwave frequency 7.49 GHz. The analyzer is based upon the quantum-number dependence of static-electric-field ionization (Ref. 5), the field ionization reducing what would be other atom ionization [by a microwave cavity (Refs. 2 and 3)]. Calibration of the analyzer is by laser excitation of the different states with the waveguide microwaves turned off. The calibration curves obeyed an n^4F_A scaling law, where F_A is the analyzer electric field strength. A small shift in the direction of more negative n_e and increasing with microwave power is due to about 0.1% of the microwave power escaping through the waveguide beam-exit aperture and entering the analyzer region. Atoms with $n = 60$ and $n_e = +59$ would ionize at about 2.2 times the field strength indicated for $n_e = -59$.

tion probability. At 0.5 W resonant transitions to $n = 59$ and to $n = 62$ also occur, along with other transitions to $n = 62$ in the region 7.0–7.2 GHz that are strong at the $n = 60$ to $n = 61$ resonance frequency. Higher-order n changing and ionization begin to occur at some frequencies for powers between 0.5 and 0.7 W ($F_\mu = 14.5$ V/cm). Experiments with the second laser transition off resonance showed that these n-changing signals were not due to direct laser coupling with $n \neq 60$ microwave sidebands.[3]

Our evidence for the n-changing behavior just described is contained in reproducible quantum-number analyzer scans for forty different microwave frequencies and various power levels. Some data for our 6.7–6.9-GHz "diffusive" region and for two frequencies outside this region are displayed in Fig. 3. We have observed no up n changing at 5.96 GHz and no down n changing in the range 6.7–7.1 GHz, although both occur simultaneously above 7.4 GHz.

In Fig. 3 no transitions occur for $F_\mu < 5$ V/cm when $F_s = 5.5$ V/cm. We have repeated our measurements at $F_s = 8.8$ V/cm and find some $\Delta n = +1$ transitions occurring at lower values of F_μ; otherwise the Stark-shift–corrected data for the two different static-field values are very similar, and are characteristic of the microwave field strength alone. Data at $n = 63$ and $F_s = 8.0$ V/cm are similar to the above and again show no discontinuities in n-changing probabilities when $F_\mu = F_s$.

The onset of diffusive n changing is determined by a bottleneck transition, here possibly $n = 61$ to 62 at 6.75 GHz. Once the resonance for the bottleneck

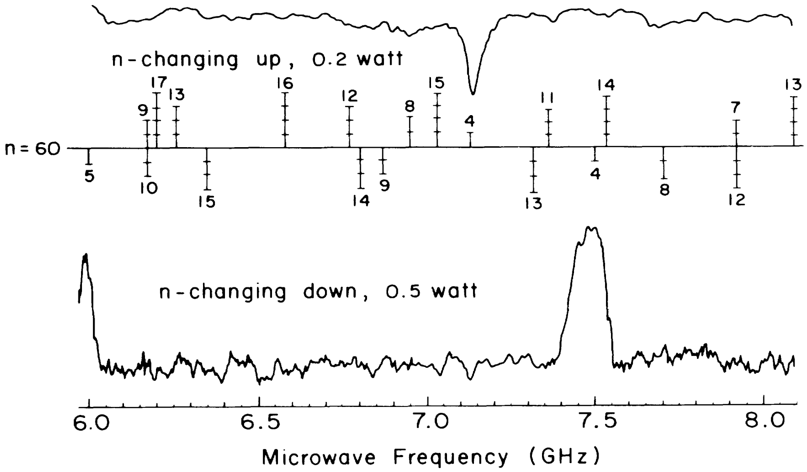

FIG. 2. The microwave frequency dependence of the n-changing signals at low microwave power, where n changes up or down only by 1. Resonant multiphoton transitions are observed near the expected static-field Stark-shifted frequencies indicated. These resonances involve the absorption of $k = 4$ or 5 microwave photons. The down n-changing atom production curve was obtained with the state analyzer field F_A set at 50.0 V/cm, while up n changing was studied as $n = 60$ atom loss with $F_A = 45.5$ V/cm (see Fig. 1). The locations of resonances for larger direct (not stepwise) changes in n are indicated along with their order k.

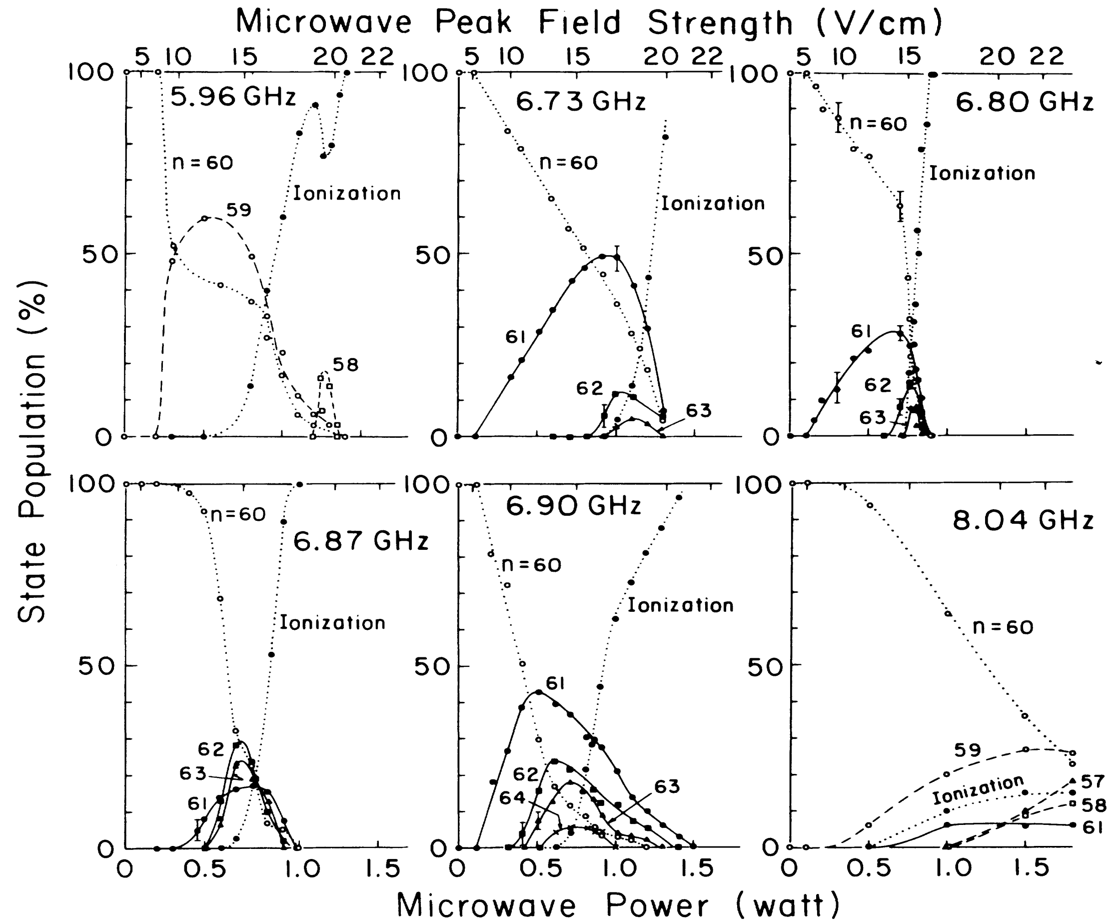

FIG. 3. Full state-analysis curves to high microwave powers for a few microwave frequencies. Microwave field dependences for n changing both up and down are shown along with those for ionization. Error bars indicate reproducibility.

transition has a width sufficient for overlap with resonances for steps to still higher states, essentially nonresonant many-state n changing would set in for some frequency band. A signature for such diffusion is a smooth distribution of product excited states, as seen in Fig. 3 notably at 6.90 GHz. The experimental peak microwave field strength at 0.8 W was 15.5 V/cm, whereas the lowest estimate for the threshold for diffusion at four times higher frequencies and $n=60$ is 5 V/cm.[1] Whether or not a threshold field for stochastic behavior has actually been reached near 6.80 GHz awaits further study.

In perturbative multiphoton absorption theory the basic expansion parameters are $(z_{11}-z_{22})F_\mu/\omega$ and $z_{12}F_\mu/\omega$, where the z_{ij} are electric-dipole matrix elements coupling Stark states.[6] At $F_\mu=10$ V/cm and for two states of adjacent n and minimal n_e, $z_{11}F_\mu$, $(z_{11}-z_{22})F_\mu$, and $z_{12}F_\mu$ are 60, 1, and 12 GHz,[7] respectively. Thus our transitions are controlled by a coupling z_{12} that decreases as the $2-\delta$ power of n, δ being a change in the final state n_e away from $1-n$.[7] This consequence of decreasing wave-function overlap underlies the observed near one-dimensionality. Our situation is the reverse of that for nonhydrogenic atoms in states of low m, where the radiative coupling z_{12} has been ignored[8] when it is smaller than the electrostatic coupling of a Rydberg electron to the core electrons. In our case the static-field level crossings that are associated with significant values of z_{12} lie at field strengths far above that for ionization and seem to play no special role.

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