Observation of Resonances in the Microwave-Stimulated Multiphoton Excitation and Ionization of Highly Excited Hydrogen Atoms[(a)]

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The multiphoton ionization of highly excited hydrogen ($45 \leq n \leq 57$) for frequencies $9.4 \leq \nu \leq 11.6$ GHz is of such high order $88 \leq k_0 \leq 173$ that quantal tunneling may compete. Nevertheless, we observe resonances in the frequency dependence of the ionization rate caused by strong multiphoton transitions of order $3 \leq k' \leq 7$ between adjacent energy levels. We show that these multiphoton transitions proceed at rates comparable to the overall ionization rate. These results rule out tunneling as the dominant intense-field ionization mechanism.

The infrared laser excitation and dissociation of molecules has been described recently as involving multiphoton transitions up a portion of the vibrational-energy-level spectrum to a quasicontinuum strongly coupled to dissociative states.[1] When the interaction dipole matrix element μF becomes comparable to the anharmonic defect A, the first transition is laser-power broadened enough for a chain of transitions to occur at "red-shifted" frequencies resonant with transitions higher up the vibrational ladder.[2,3] The experimental observation of this theoretically predicted effect in molecules may be obscured by the rotational-energy-level substructure, however, with an appropriate set of rotational levels supplying a different way to compensate for the vibrational-state anharmonicity.[4] The work in molecules has been limited to relatively weak fields coupling intermediate energy levels in the ladder via one-photon transitions rather than multiphoton transitions.

In this Letter we present experimental evidence that resonant microwave *multiphoton* transitions up one or more steps on the local ladder of highly excited hydrogen atomic levels can be stimulated and are often rapid enough to enhance strongly the microwave atomic ionization rate. The quasicontinuum cutting the top of the hydrogenic spectrum consists of those higher ionizing levels very strongly coupled to the continuum by the oscillating electric field $F = F_0 \cos\omega t$. The strong-field nature of our experiments on atoms with principal quantum number n near 50 can be specified by noting that for a typical case of $n_1 - n_2 \sim \frac{1}{2}n \sim 25$ and $F_0 \sim 75$ V/cm, the peak first-order Stark energy $\mu F_0 = \frac{3}{2}n(n_1 - n_2)F_0$ in atomic units[5] was about 150 GHz, larger than the classical electron orbital frequency, 50 GHz, the microwave frequency, 10 GHz, and the local effective anharmonic defect, 3 GHz, respectively. These conditions may produce appreciable population of ac-Stark-effect "sideband" energy levels.[6]

The experiments on microwave ionization began with the formation of an atomic beam containing highly excited atoms selectively laser-excited into an individual n level by resonant one-photon absorption. This beam then passed through a microwave field of appropriate field strength where a fraction of the atoms was ionized. The resultant protons in the beam were charge selected and counted with an electron multiplier. The laser-beam intensity was chopped for phase-sensitive detection of the signal protons.

A second series of experiments investigated microwave-induced multiphoton transitions from the initial laser-pumped highly excited state to higher-lying bound states. Detection of such transitions involved ionization by a subsequent second weaker microwave field.

Our studies employed fast-beam Rydberg atom production and detection techniques developed previously.[7-12] A mixed excited-state hydrogen-atom beam with an energy near 10 keV was produced from a proton beam by electron-transfer collisions, usually $H^+ + Xe \rightarrow H + Xe^+$. The population of a mixture of Stark sublevels of an individual state with principal quantum number $n = n_0$ was then enhanced by directing cw-laser photons parallel to the atomic beam to induce transitions from a certain lower-lying level. An ultraviolet argon-ion laser pumped the $2s$ state,[7] while, alternatively, a CO_2 laser pumped the $n = 10$ state.[9] The atom velocity was set to Doppler tune the fixed-frequency laser radiation into resonance.[7-10]

The multiphoton ionization rate was measured as a function of microwave frequency swept continuously in a TE_{10} wave guide over a frequency range from 9.4 to 11.6 GHz. The atoms were ex-

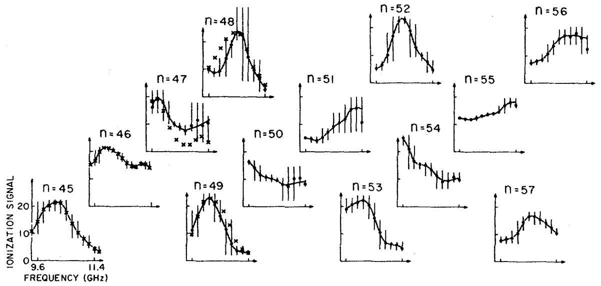

FIG. 1. The microwave ionization of highly excited hydrogen atoms as a function of frequency between 9.6 and 11.4 GHz, for each principal quantum number n_0 between 45 and 57. The vertical scales are relative units different for each n_0. The meaning of the error bars is discussed in the text. Data points indicated by crosses and dots were obtained with atoms pumped by CO_2 laser and ultraviolet argon-ion laser, respectively.

posed to ~10^2 cycles of the microwave electric field which was polarized along the atomic-beam direction. The microwave source consisted of a cw solid-state sweep generator driving a 20-W traveling-wave-tube amplifier. Since the ionization rate depended strongly upon the field strength, a great deal of effort was put into making a transmitted-power–leveling feedback loop containing a *p-i-n*-diode modulator be as frequency independent as possible.

Typical ionization signals are shown in Fig. 1 for each n_0 in the range $45 \leqslant n_0 \leqslant 57$. The data shown were obtained by smoothing once[13] the raw signal counts stored in 11 to 13 multichannel scaler bins swept synchronously with the microwave frequency.

The fluctuations in ionization signals caused by imperfect microwave-power leveling were observed in additional experiments by storing signal counts in a hundred bins. The error bars shown in Fig. 1 are local upper bounds on such fluctuations. We emphasize that spectra for adjacent values of n_0 could be taken under constant conditions for the entire microwave system and laser system; only the value of n_0 was changed by a small shift (usually ~2%) in the atomic-beam velocity. To cover the entire range of n_0, however, it was necessary to readjust empirically the leveled power to produce ionization probabilities in the range 1–30%. The equation $F^2 = -542n_0 + 32\,193$ (V/cm)^2 approximately described the variation of field strength with n_0.

The unmistakable resonant structure displayed in Fig. 1 was not strongly dependent either on the exact base microwave-power level used or on the choice of laser used to prepare the highly excited

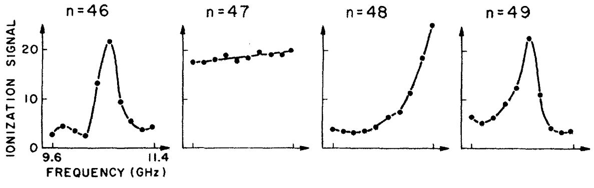

FIG. 2. Microwave multiphoton excitation signals (double-field data) for transitions from $n = n_0$ to $n \geqslant n_0 + 1$ as a function of frequency between 9.6 and 11.4 GHz for CO_2-laser–excited initial states with n_0 between 46 and 49.

beam. The observed resonance frequencies shift to lower frequencies as n_0 is increased, with "new" resonances appearing at higher frequencies as the "old" ones drop below 9.6 GHz. An interpretation of the resonance frequencies in terms of the field-free spectrum of the hydrogen atom is possible. Let k' be the number of microwave photons needed to cause a transition between field-free levels n_0 and $n_0 + 1$, that is, the first step up the ladder to the continuum. For the higher values of n_0, $k' = 3$ is in the present frequency range; the frequencies for $k' = 2$ and 4 are far away. The measured resonance frequencies of Fig. 1 are red-shifted down by about 5–10% from the lowest value of k' near our frequency range in all cases except $n_0 = 45$. There a 10% red-shifted $k' = 6$ resonance lies near an unshifted one with $k' = 7$, and the situation is more complicated.

The results of the second type of experiment designed to observe directly transitions induced by the wave-guide field from the level n_0 to levels with $n > n_0$ give more information on the multiphoton transitions up the first steps of the ladder toward ionization. After passing through the wave guide the atom beam was sent through a second microwave field inside a TM_{010} cavity resonating at 9.91 GHz. Its electric field oriented along the beam direction was set empirically to a value that produced a small ionization probability for the laser-excited initial state n_0. The important feature of the new "two-field" experiments was to voltage label[8,11] the cavity rather than the wave guide. Thus only ionizable atoms leaving the frequency-swept guide that were subsequently ionized in the cavity could contribute to our signal. The wave-guide field was maintained at the same value as that producing the ionization of Fig. 1. If it also induced transitions to bound levels with $n > n_0$, then the cavity would ionize the more highly excited atoms and produce signals. Such signals were indeed found and depended critically upon n_0, and the frequency and strength of each

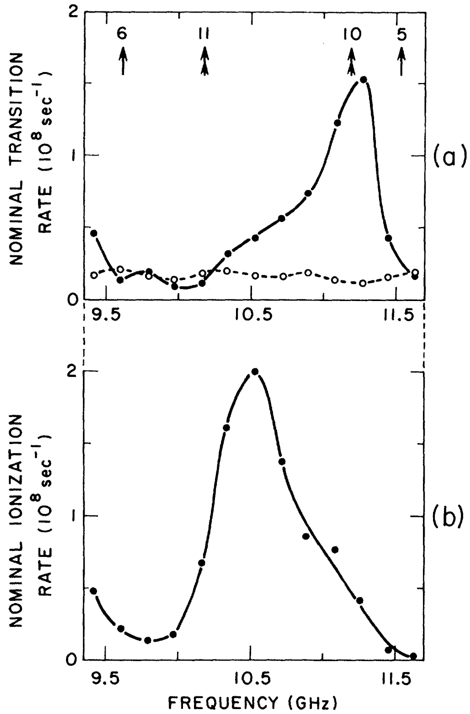

FIG. 3. A detailed comparison of the frequency dependence and absolute rates for (a) excitation and (b) ionization with $n_0 = 48$. The open circles shown in (a) are the background signals with the wave-guide field turned off. Single- and double-headed arrows show the position of multiphoton transition frequencies between a field-free $n_0 = 48$ state and field-free $n = 49$ and $n = 50$ states, respectively, for the number of photons shown.

field.

The unsmoothed results of a number of two-field studies of bound-bound multiphoton transitions are shown in Fig. 2. Quantitative data taken under identical atomic-beam, laser, and wave-guide-field conditions (peak $F = 80$ V/cm) for both the one-field ionization and the two-field excitation experiment are compared for $n_0 = 48$ in Fig. 3. Absolute scales for both rates were determined from the observation of experimental saturation of signals with wave-guide power.

The resonances observed in the signals for bound-bound transitions have maxima usually lying close to some predicted frequencies for multiphoton transitions with $\Delta n = 2$, ignoring ac Stark shifts. These resonant signals could contain contributions from $\Delta n \geq 2$ transitions, but detection of $\Delta n = 1$ signals were also possible for the cavity-field strengths employed. In fact, it is quite possible for these signals to be due primarily to $\Delta n = 1$ transitions that are ac Stark shifted about 3% below field-free frequencies. The Stark shifts range from 250 MHz at $n_0 = 49$ ($k' = 5$) to 350 MHz at $n_0 = 45$ ($k' = 7$). As explained above, the ionization resonances are shifted about 10%. The bound-bound resonances were far less sensitive to the wave-guide-field fluctuations with frequency, as might be expected. Note that the peak rates for the excitation and ionization processes are comparable, and that which rate is higher depends on the frequency.

Comparisons of bound-bound and bound-free rates such as those in Fig. 3 provide considerable evidence for a major ionization mechanism involving chains of multiphoton transitions enhanced by the upper bound states of the atom. When compared to one another and to the local level anharmonicity, the frequencies of the two kinds of resonance maxima strongly support this conclusion, as do the comparable magnitudes for the rates. Our previous ionization experiments[11,12,7] displayed a gross frequency dependence in qualitative agreement with the near-adiabatic (modified tunneling) behavior predicted by a certain class of untested theories of nonresonant high-order ionization.[14] This region is characterized by the free electron energy in the oscillating field being at least comparable to the atomic ionization potential, a condition satisfied in our experiments. However, the presently observed resonant ionization phenomena are not explained in terms of a direct tunneling process, even including the possible effects of crossing of Stark energy levels.[15] The observed intense-field resonant enhancement of the ionization arises because the peak dipole-interaction energy is at least comparable to the photon energy and approaches the local energy-level separation of the field-free atom. Such effects can be described by a physical picture that includes saturation of those multiphoton absorption processes included in the development of oscillating hydrogenic Stark states.[6,16,17]

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Impact-Parameter Dependence of K-Shell Vacancy Sharing in Heavy-Ion Collisions

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The impact-parameter dependence of the $2p\sigma$-$1s\sigma$ vacancy-sharing probability has been investigated in 35-MeV Cl on Ti and Cl on Ni collisions. A stronger impact-parameter depencence is observed with increasing asymmetry of the collision system. The impact-parameter dependence of the K-vacancy sharing is in good agreement with a simple analytical solution, derived by Briggs, of the two-state coupled-channel equations.

In nearly symmetric heavy-ion collisions, the distribution of K-shell vacancies between projectile and target is well described in terms of the vacancy-sharing process.[1] It is assumed that the K vacancies in the higher-Z collision partner are only produced by vacancy transfer from the $2p\sigma$ to the $1s\sigma$ molecular orbitals (MO) via radial coupling, in the outgoing half of the trajectory. The vacancy-sharing probability was calculated by Meyerhof[1] using the charge-transfer model of Demkov[2] and good agreement was obtained with experimental results of total cross sections.[1,3] However, the probability was assumed to be independent of the impact parameter b. Briggs[4] has recently calculated the b dependence of the vacancy-sharing probability. According to his calculations a pronounced b dependence is expected for asymmetric collision systems which has not been experimentally tested up to now. In this Letter we present experimental evidence for the impact-parameter dependence of the K-vacancy-sharing probability.

The K–x-ray emission probabilities for the projectile and the target atoms have been simultaneously measured in 35-MeV Cl on Ti and Cl on Ni collisions. The experiments were performed at the EN tandem accelerator of the Max-Planck-Institut für Kernphysik at Heidelberg. The targets