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Stark-induced resonances in the photoionization of hydrogen

Wallace L. Glab and Munir H. Nayfeh
Department of Physics, University of Illinois at Urbana-Champaign, 1110 W. Green Street, Urbana, Illinois 61801
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We report the first observation of electric-field-induced resonances in the multistep photoionization of hydrogen. Asymmetric profiles having blue wings are observed near the photoionization limit resulting from tunneling across the Stark-Coulomb combined potential (shape resonance). We measured the dependence of the spacing of the resonances on the electric field strength.

Considerable experimental and theoretical efforts have been recently undertaken to study the photoionization spectrum of atoms in the presence of external electric fields whose interaction energies are comparable to the Coulomb energy.[1–13] In the region of the photoionization threshold calculations predict the presence of a set of discrete levels with short but finite lifetimes, giving rise to a series of broad resonances in the optical absorption which extend past the zero-field ionization limit.[6–9]

The simplest and most abundant atom in nature, the hydrogen atom, has been the subject of continuing interest to atomic physicists. This interest stems from the fact that the absence of core effects allows precise calculations to be made of its properties, both in isolation and under the influence of external forces due to electromagnetic fields and collisions. For an atom in an external electric field, the electron-core interaction makes the otherwise separable Schrödinger equation nonseparable and hence less amenable to theoretical calculations. Thus, the study of atomic hydrogen presents the opportunity for a detailed comparison of theory and experiment in situations where theoretical descriptions of complex atoms are absent or imprecise.

So far all reported experiments have been on complex atoms in spite of the fact that there are numerous ongoing efforts to study this effect in hydrogen. In this Rapid Communication we report the first observation of Stark-field-induced modulation in the photoionization spectrum of atomic hydrogen, which revealed asymmetric profiles having blue wings near the photoionization limit due to tunneling across the potential barrier of the combined Coulomb and Stark fields. Because this interaction has been calculated very accurately in hydrogen, this experiment allows, for the first time, comparisons between theory and experiment for hydrogen.

A number of methods for the excitation of hydrogen Rydberg states have been employed by various research groups, which include charge exchange,[14] Doppler tuning of a relativistic atomic hydrogen beam crossed with a powerful, nontunable laser,[15] simultaneous absorption of three photons near 273 nm,[16] and excitation of the Lyman α transition with laser radiation at 122 nm, followed by excitation to Rydberg states with a second laser at about 365 nm.[17]

In here we use a technique which has already been tested in our laboratory, which circumvents some drawbacks of the methods described above.[18] Simultaneous absorption of two photons from a single tunable pulsed laser beam at 243 nm results in excitation from $1s$ to $n=2$, and some photoionization of the resulting $n=2$ population. A second pulsed beam excites states near the continuum from the $n=2$ state. For properly chosen 243-nm energy densities, a large population of $n=2$ atoms can be produced with only a fraction of a percent of them being photoionized. The second beam is then capable of promoting a large portion of the remaining excited atoms to a well-defined high-lying state without saturating the process. Previously two-photon Doppler-free spectroscopy of a $n=2$ state has been performed using radiation at 2430 Å; however, no attempt to excite the hydrogen atoms further was reported.[19]

Figure 1 shows a block diagram of the experimental setup. The atomic hydrogen source is modified Wood discharge tube.[20] An atomic beam is formed by effusion from the discharge region through a multicollimator assembly composed of 25 small glass capillaries. The thin-walled capillaries are 4 mm long, with an inside diameter of about 0.2 mm. The resulting atomic beam is directed into the diffusion pumped cell which contains the field plates. The beam is loosely collimated, but produces a density of about 10^{11} H^0/cm^3; the background gas density is on the order of $10^{12}/cm^3$. The electric field between the plates is determined to an accuracy of 0.5% by measurement of the applied voltage and the separation of the field plates. Ions produced by the laser radiations are driven by the electric field through a grid in the grounded plate. They travel

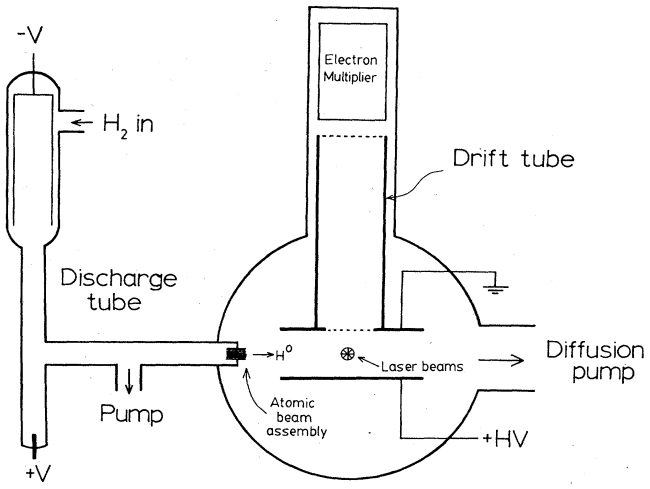

FIG. 1. Block diagram of the experimental setup.

through a 45-cm-long, field-free drift tube which provides mass analysis. This is necessary since molecular impurities are easily ionized by the ultraviolet wavelengths in use. At low electric fields, the mass resolution is sufficient to verify that the signal under study is indeed a result of atomic hydrogen. The resolution at kV/cm range fields is sufficient to separate the hydrogen signal from those due to the impurities. Ions are detected using an 18-stage venetian blind electron multiplier capable of single-ion detection. Under typical experimental conditions, several hundred ions are detected per pulse.

The optical beams needed for the excitation of atomic hydrogen are produced using a pulsed laser system: an Nd^{3+}:yttrium aluminum garnet (YAG) laser and two dye lasers. A fraction of the second harmonic of the YAG laser at 532 nm is used to pump one of the dye lasers producing a beam at 630 nm, which is frequency doubled to 315 nm by a potassium dihydrogen phosphate KH_2PO_4 (KDP) crystal and then summed with the residual YAG fundamental by a KDP crystal resulting in a beam of 243 nm of pulse length of about 10 ns, a bandwidth of about 1.5 cm^{-1} and pulse energies on the order of 10 μJ. The second dye laser produces a beam at about 555 nm which is summed with part of the YAG fundamental to produce a beam with pulse length near 10 ns, bandwidth of 0.6 cm^{-1}, pulse energies of a few tenths of a millijoule and a wavelength near 365 nm.

The data are collected and analyzed using a Digital Equipment Corporation LSI-11 computer system. Peak detectors sample and hold the intensities and signal for each beam, and analog-to-digital converters digitize the voltage for storage. Several hundred pulses are stored for each wavelength of the second beam; subsequently, a program removes the effects of intensity variations. The result of a complete data run is then the laser-induced ionization as a function of wavelength for a given pair of beam intensities.

Calibration of the wavelength of the mixed beam which drives the photoionization is performed through the use of the optogalvanic effect using a commerical He-Ne laser discharge tube. Comparison of the positions of several He resonances to the dye laser wavelength counter allows us to calibrate the wavelength scale of the electric field study scans to within ~ 0.5 cm^{-1}.

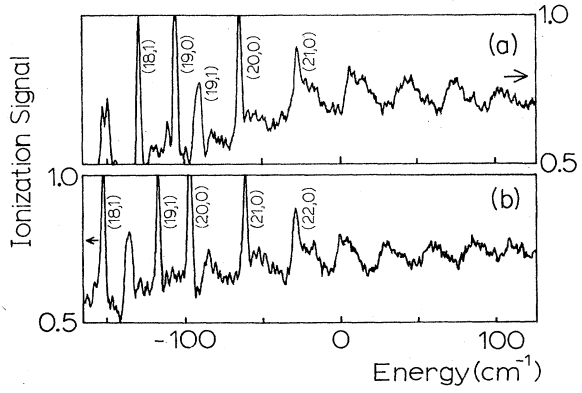

FIG. 2. Photoionization spectrum of hydrogen near the field free photoinozation threshold $E=0$ in the presence of two different Stark fields (a) 8.0, and (b) 6.5 kV/cm. In (a) and (b) both light beams have π polarization. The arrows point to the appropriate scales. Some quasidiscrete states are labeled by (n_1,n_2). All states are $m_l=0$.

Figures 2(a) and 2(b) show the photoionization spectrum near $E=0$ at two values of electric field amplitudes, and light polarization along the field (π polarization). Below $E=0$, there are sharp symmetric peaks which correspond to the highly excited quasidiscrete Stark states. The peak assignments are made by comparison to numerical calculations.[20] As $E=0$ is approached, fairly broad resonances appear starting just below $E=0$ and persisting into positive energies. These resonances show significant degrees of asymmetry in the form of blue wings in the region near $E=0$. The depth of these modulations near $E=0$ is measured to be $\sim 18\%$ of the underlying smooth photoionization yield at 8 kV/cm.

It was noted, that in nonhydrogenic systems, core effects may result in asymmetric profiles for resonances below $E=0$.[2] In a recent paper, these effects were studied in detail for closed shell cores using the WKB approximation.[8] The calculation showed that interference effects between continuous and quasidiscrete channels that are coupled by the nonhydrogenic core induce significant asymmetry which would not be present in hydrogen. In the region above $E=0$, these interference effects do not play any role. However, the question whether even in this region the profiles may show asymmetry was raised previously.[6] In fact, it was noted that the process of coupling the ξ and η motions has resemblances to autoionization without any core interactions, since it provides coupling of bound states to underlying continua. Recent theoretical calculations established that tunneling may produce an asymmetry due to different tunneling rates below and above a quasiresonant state (shape resonance). Although this symmetry occurs in hydrogen as well as in complex atoms, the core modulates it further.[7-8,10]

A detailed photoionization spectrum of hydrogen $3p$ state was calculated using WKB theory at field amplitude which is much higher than the experimental field (77 kV)/cm.[8] Although direct quantitative comparison with this theory is not possible at this time because of this and because s and p states are involved in our experiment, the above outlined features of the spectra have some resemblance to those at higher fields. Specifically the calculation predicts the presence of asymmetries in the profiles near $E=0$ resulting from tunneling. Such tunneling is not observable in complex atoms because it is masked by the electron-core effects. Moreover, our preliminary measurements of the spacing of the broad resonances at threshold show that it agrees with the prediction of the theory.

In the study of the polarization properties of the Stark-induced levels near the photoionization threshold, one needs to consider the effect of the E field on the intermediate state $n=2$.[22,23] In the absence of the field, the two-photon excitation of $1s_{1/2}-2s_{1/2}$ with π polarization does produce a pure $m_j=0$ state. At 475 V/cm, the Stark effect and the Lamb shift become comparable, where as at ~ 2.910 kV/cm the Stark effect becomes comparable to the fine-structure splitting. For fields greater than about 5 kV/cm, the interaction with the Stark field dominates over the fine structure, thus leading to a linear Stark splitting of $n=2$. In the region of 2–5 kV/cm (field strengths suitable for observation of Stark-induced levels in $E \geq 0$ region), the calculations of the $n=2$ Stark effect are quite complicated. Only n and m_j are good quantum numbers, but neither

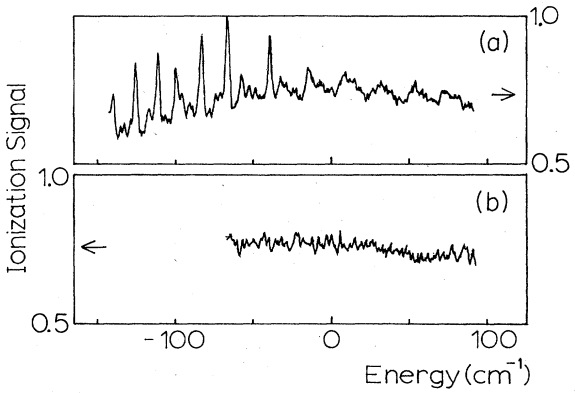

FIG. 3. Photoionization spectrum of hydrogen near the photoionization threshold $E=0$ in the presence of 4.5 kV/cm. In (a) both light beams have π polarization and in (b) the exciting and ionizing beams are of π and σ polarization, respectively.

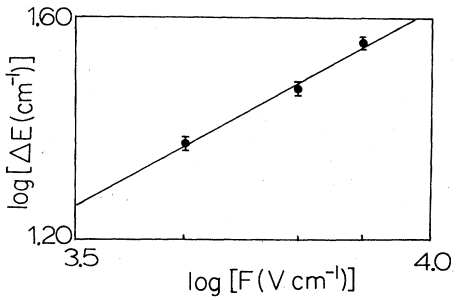

FIG. 4. Dependence of the spacing of Stark-modulated spectrum near $E=0$ on the Stark field. This plot of log of spacing vs log of the field also shows a least-squares fit of the data points giving a slope of 0.7 ± 0.07.

j and l nor the parabolic quantum numbers are. Such calculations were previously done for $n=2$, 3, and 4.[22] Using these results, one can show that the fraction of $n=2$, $m_l=0$ produced in our two-photon scheme is 0.8 and 0.9 at 1.0, and 4.5 kV/cm, respectively.

We should mention that the $n=2$ state in the presence of the electric field has two $m_l=0$ states. One of them is blue shifted and the other is red shifted. At the field strengths we are using the splittings of these two states is less than the effective bandwidth of the laser. Therefore, both of these states are populated by our two photon process.

The corresponding fractions of $|m_l|=1$ produced are 0.2 and 0.1. If the ionizing pulse has σ polarization, then one expects to excite $m_l=0$ and $|m_l|=1$ and $|m_l|=2$ final Stark-induced resonances. Only 50% of these fractions, i.e., 0.1 and 0.05 are $m_l=0$. Hence, one expects essentially to see no modulation in the range 2–5 kV when a σ-polarized ionizing laser is used. We took some measurements of the polarization spectrum using π- and σ-polarized ionizing laser radiation at 4500 V/cm. The results, given in Figs. 3(a) and 3(b), respectively, confirm the above estimates.

We studied the field dependence of the spacing. Figure 4 shows the separation at threshold as a function of the electric field strength. Although our results which give a slope of 0.7 ± 0.07 show that the spacing can be described by a $\frac{3}{4}$ power law as predicted by the same WKB theory, the accuracy of the data is not sufficient to rule out a somewhat different power law.

In conclusion, we have observed asymmetric profiles having blue wings near $E=0$ resulting from tunneling across the Stark-Coulomb barrier and measured the field dependence of their spacings. Comparison between the effect in hydrogen and those in potassium and krypton is underway to study the role of the core on the resonances.[12]

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