tion and comparisons between homologous atoms, to evaluate consistent f values for transitions in ions with higher charge states than those studied experimentally in the present investigation.

ACKNOWLEDGMENT

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Multiphoton Ionization of Atomic and Molecular Hydrogen at 0.53 μ

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This work reports multiphoton ionization and dissociation of molecular hydrogen under the influence of laser radiation at 0.53 μ. The six-photon ionization probability of hydrogen atoms is derived for laser radiation acting upon atomic hydrogen from a molecular hydrogen beam which is up to 80% dissociated by radio-frequency power. The experimental six-photon ionization probability of atomic hydrogen has been found to be in good agreement with the corresponding probability calculated using time-dependent perturbation theory if the statistical properties of the incident light are taken into account.

I. INTRODUCTION

The nonlinear interaction between an intense beam of photons generated by a Q-switched laser and rare gases or alkali-metal atoms at low pressure leads to ionization of these atoms through the simultaneous absorption of several quanta if the laser radiation is sufficiently intense.[1-5] Multiphoton-ionization processes have been the subject of a considerable amount of recent theoretical work, especially within the framework of time-dependent perturbation theory.[6-11] The simultaneous absorption of K_0 photons by an atom corresponds to a direct transition between the ground state and the continuum, K_0 being the next integer greater than the ionization energy E_i of the atom divided by the photon energy E_p. The multiphoton-ionization probability W is proportional to Γ^{K_0},

$$W = \alpha \Gamma^{K_0} \ , \tag{1.1}$$

where the laser intensity Γ is expressed in photons $\mathrm{cm}^{-2}\,\mathrm{sec}^{-1}$ and α is a factor depending on the atom considered and of the photon energy E_p.

Experimental results of the multiphoton ionization of atoms with a doped Nd-glass laser show that the multiphoton-ionization probability is proportional to the laser intensity Γ raised to a power K somewhat less than the K_0 value when there is a quasiresonance between the energy of an atomic level and the energy of the integral number of photons closest to the K value. When such a quasiresonance occurs, the atom appears to absorb K photons corresponding to the transition between the ground state and an excited state; the atom then absorbs $K_0 - K$ additional photons corresponding to the $K_0 - K$ photons ionization from this excited state. The probability of this latter transition is much greater than the K-photon excitation probability for the laser intensity used. Under these conditions, the multiphoton-ionization probability would be essentially determined by the K-photon process, and it is therefore of no value to compare a calculated K_0-photon-ionization probability to a measured K-photon-ionization probability.

It is better to experiment with atomic hydrogen than with other gases for two reasons: First, theoretical calculations lead to accurate multiphoton-ionization probability values because of exact knowledge of wave function; most theoretical works concern atomic hydrogen. Second, the levels of atomic hydrogen are so few that there is no possible quasiresonance at the wavelength 0.53 μ, the second-harmonic generation of the Nd-glass laser.

The purpose of the present paper is to describe an experiment in which the six-photon-ionization probability of atomic hydrogen has been measured at 0.53 μ. It is of special interest to compare this value with the corresponding calculated six-photon-ionization probability. This is the first time to our knowledge that such a comparison has been done for atomic hydrogen.

II. EXPERIMENTAL ARRANGEMENT

The Nd-glass laser used is Q switched by a rotating prism. The maximum peak power delivered is 1.5 GW distributed over many modes. After frequency doubling in a potassium dihydrogen phosphate (KDP) crystal the second-harmonic power available is 200 MW at 0.53 μ. Figure 1 shows the experimental arrangement. The green laser radiation is focused into a vacuum chamber with an aspheric lens L_1 (focal length 50 mm). This lens can be moved along the laser axis. The residual pressure in the experimental chamber is less than 10^{-7} Torr. The laser intensity is not uniform at the focus of the lens L_1 and may be expressed in the form

$$\Gamma(x,\ y,\ z,\ t) = \Gamma_0 F(x,\ y,\ z) G(t) \ , \tag{2.1}$$

where Γ_0 is the maximum intensity, $F(x,\ y,\ z)$ is the normalized function representing the spatial intensity distribution, and $G(t)$ is the normalized function representing the temporal intensity distribution. This expression is strictly valid for a single-transverse-mode laser beam only as the spatial intensity distribution could vary during the duration of the laser pulse if laser emission takes place into several transverse modes. However, by using a TRW streak camera, we verified that no significant variation in the spatial intensity distribution occurred during the duration of the laser pulse when an aperture was included in the laser beam. Γ_0 and $F(x,\ y,\ z)$ are measured photometrically,[1] whereas $G(t)$ is determined from an oscillogram of the laser pulse. The following definitions are made. The effective interaction volume for a Kth-order process is

$$V_K = \iiint F^K(x,\ y,\ z)\, dv \ . \tag{2.2}$$

Thus, at 0.53 μ, $V_6 = (1 \pm 0.4) \times 10^{-7}\ \mathrm{cm}^3$. The effective interaction time for a Kth-order process is

$$\tau_K = \int G^K(t)\, dt \ . \tag{2.3}$$

Thus, at 0.53 μ, $\tau_6 = 1.1 \times 10^{-8}$ sec. The multiphoton-ionization probability is then

$$W = N_i / n_0 V_K \tau_K \ , \tag{2.4}$$

where N_i is the number of hydrogen ions produced and n_0 is the density of hydrogen atoms.

Atomic hydrogen is produced from a molecular hydrogen beam which can be partially dissociated

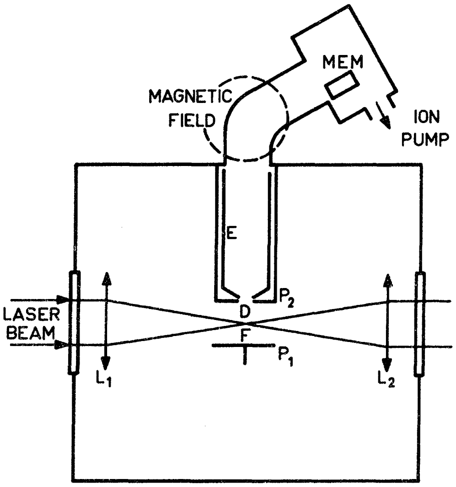

FIG. 1. Schematic diagram of the experimental arrangement.

by rf power before the interaction with the laser radiation. Molecular hydrogen with a small admixture of water vapor flows through a glass tube which terminates with a capillary of 1 cm length and 1 mm diam. The axis of the beam is perpendicular to both the laser axis and ion detection axis. The end of the capillary tube is located 2 cm from the focus of the lens L_1, where the molecular hydrogen density is 10^{12} cm^{-3}. The thermal dissociation energy of the H_2 molecule is 4.5 eV. This energy is supplied by a rf generator delivering up to 200 W at 260 MHz. The rf generator is inductively coupled to the glass tube through which molecular hydrogen flows. The density of the hydrogen atoms resulting from thermal dissociation alone has been determined with the laser off. A small platinum plate (diameter 5 mm) located 2 cm from the end of the capillary tube is heated by the recombination of hydrogen atoms to form a molecule on its surface. The dissociation efficiency is a function of rf power, and reaches 80% for a molecular hydrogen pressure of 10^{-1} Torr up-stream in the capillary tube.

The atomic H^+ and molecular H_2^+ ions resulting from the laser interaction at the focal point are analyzed with a magnetic mass spectrometer. Ions are extracted with a transverse electric field (1500 $V cm^{-1}$) applied between plates P_1 and P_2. These ions pass through a circular aperture D (diameter 4 mm), a focussing ion-optical system E biased at 150 V, are mass separated in a magnetic spectrograph with a variable magnetic field (maximum 2000 G), and are then detected with a magnetic electron multiplier MEM Bendix type 308. An ion pump maintains the pressure at less than 10^{-5} Torr in the region of the electron multiplier when hydrogen is admitted into the experimental chamber.

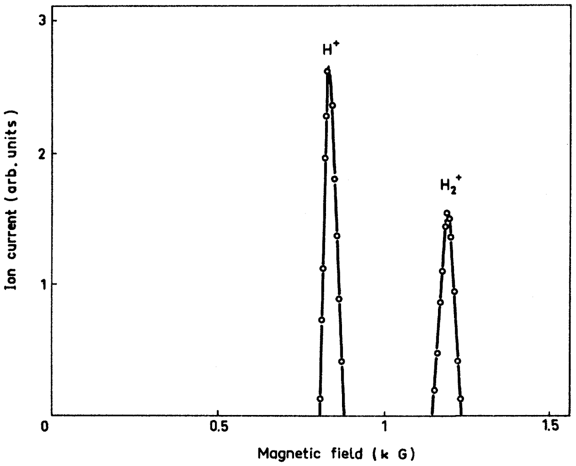

FIG. 2. Typical mass spectrum of H^+ and H_2^+ obtained for a laser intensity $\Gamma = 3 \times 10^{12}$ W/cm^2.

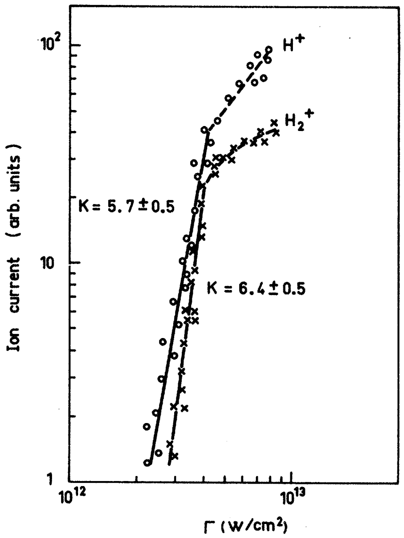

FIG. 3. Log-log plot of H^+ and H_2^+ from H_2 as a function of the laser intensity Γ.

III. INTERACTION OF 0.53-μ LASER RADIATION WITH MOLECULAR HYDROGEN

A. Experimental Results

An experiment was first conducted without dissociation of the molecular hydrogen beam by rf power. Both atomic and molecular hydrogen ions were detected. Figure 2 shows a typical mass spectrum obtained by tuning the magnetic field of the mass spectrometer for a fixed laser intensity of 3×10^{12} $W cm^{-2}$. The H^+ and H_2^+ signals both arise from the focal region and not from the lenses L_1 or L_2 nor from the plates P_1 or P_2. This was verified by moving the lenses L_1 and L_2 along the laser axis, causing the H^+ and H_2^+ signals to vanish as soon as the focal point is no longer opposite the aperture D (Fig. 1). It has also been verified that these signals do not arise from multiphoton ionization of impurities in the residual pressure. Therefore, the atomic hydrogen ions result from the interaction of the laser radiation itself with molecular hydrogen through a multiphoton process. Total dissociation of I_2 vapor has previously been observed under the influence of the output of a ruby laser[12] as well as partial dissociation of molecular hydrogen.[13] Theoretical work concerning multiphoton dissociation of molecules under the influence of laser radiation has been undertaken.[14,15]

The experiment consists of a measurement of the output current of the electron multiplier as a function of the laser intensity at the focal point. Figure 3 represents the log-log plot of the H^+ and H_2^+ signals vs the laser intensity. The slope is 5.7 ± 0.5 for H^+ and 6.4 ± 0.5 for H_2^+. To precisely correlate the electron multiplier output current

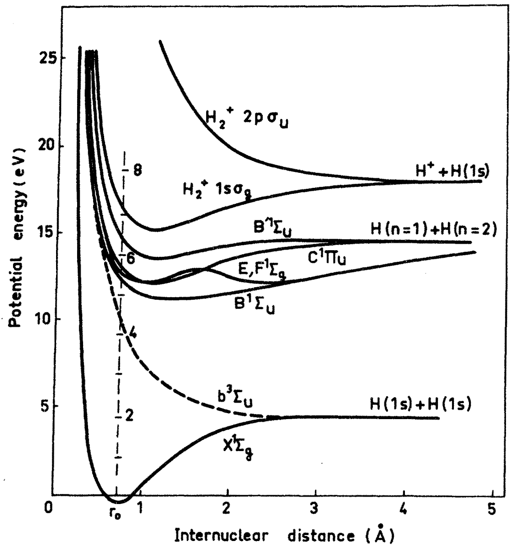

FIG. 4. Potential-energy curves for H_2 and H_2^+. The dashed line represents the multiphoton transitions from the X ($v=0$) ground state level satisfying the Franck–Condon principle.

with the number of ions produced at the focal point, previous results concerning multiphoton ionization of rare gases can be taken into account. It has been shown[1] that the law of variation of the number of ions produced vs the laser intensity changes slope when all the atoms or molecules in the interaction volume V_K are ionized. The increase in the number of ions observed beyond this saturation point would simply correspond to ionization of atoms in the neighborhood of this interaction volume V_K and the corresponding slope depends only upon the characteristics of the focusing system. In the present work, the lens has been corrected for the various aberrations and the interaction volume V_K is well defined. The discontinuity is therefore determined with a good precision and corresponds to a number of ions $N_i = n_0 V_K$. The measurement of n_0 and V_K permits the determination of the absolute value of the number of ions corresponding to the saturation point, and serves to calibrate the curves of Fig. 3.

B. Formation of the Observed Atomic Hydrogen Ions H^+

To understand the origin of the observed atomic-hydrogen ions H^+, an examination of the potential-energy curves of H_2 is necessary. The most important electronic states[16] appear in Fig. 4, where the potential energy is expressed both in eV and in number of photons. The selection rules for molecular transitions may be summarized as follows[17]: Optical transitions with spin multiplicity $\Delta S \neq 0$ are forbidden due to the weak spin-orbit interaction in H_2. Thus, the transitions from the ground state $X\,^1\Sigma_g$ (singlet) to the repulsive state $b\,^3\Sigma_u$ (triplet) are forbidden. The second rule concerns parity so that starting from the ground state $X\,^1\Sigma_g$, even order transitions are allowed to even states and odd order transitions are allowed to odd electronic states. The same rule is applied for intermediate states. Finally, the multiphoton optical transitions which occur without change in the electronic state of the molecule seem to be forbidden by the Franck–Condon principle. The latter asserts that the more probable transitions are those which take place without significant change either in relative positions of the nuclei (i.e., vertically) or in relative momentum of nuclei in the molecule. Thus, for example, the even order transitions from the initial ($v=0$) $X\,^1\Sigma_g$ level into the continuous term spectrum of the same ground X state are highly forbidden, because a very significant change in momentum would occur during such a "vertical" transition.

Under these conditions, the higher electronic states are expected to play a very important role in multiphoton dissociation processes of the hydrogen molecule. The following processes may be suggested to explain the observed slope $K=5.7$ for H^+. A six-photon transition may occur from the most populated ($v=0$, $k=1$) level of the ground state of the molecule to the ($v=21$, $k=2$) level of the double minimum E, $F\,^1\Sigma_g^+$ state,[18] that is,

$$H_2 + 6E_p \rightarrow H_2 E,\ F\,^1\Sigma_g^+\,(v=21,\ k=2). \tag{3.1}$$

Then an additional photon absorption can take place to reach the continua of the $B'\,^1\Sigma_u$ or $C\,^1\Pi_u$ states, that is,

$$H_2 E,\ F\,^1\Sigma_g^+(v=21,\ k=2) + E_p \rightarrow H(n=1) + H(n=2). \tag{3.2}$$

The atom of hydrogen with principal quantum number $n=2$ is then ionized through the simultaneous absorption of two photons. It should be pointed out that the one-photon and the two-photon transitions are saturated. In the high laser intensity used, $\Gamma = 3\times 10^{12}$ W/cm², the two-photon-ionization probability of 2s metastable hydrogen is $W_2 = 10^{13}$ sec^{-1}.[7] W_2 is so high that with $\tau_2 = 2\times 10^{-8}$ sec, $W_2\tau_2 = 2\times 10^5 \gg 1$. The total probability of obtaining an H^+ ion is then only governed by the six-photon process. The following process may also give a small contribution to the observed H^+ ions. A five-photon transition from the ($v=0$, $k=1$) level of the ground state would put the hydrogen molecule into the ($v=3$, $k=5$) level of the $B\,^1\Sigma_u$ state,[19] that is,

$$H_2 + 5E_p \rightarrow H_2 B\,^1\Sigma_u\,(v=3,\ k=5)\ . \tag{3.3}$$

Then an additional absorption of two photons would dissociate the molecule into the $B'\,^1\Sigma_u$ and $C\,^1\Pi_u$ states continua, that is,

$$H_2 B\,{}^1\Sigma_u(v=3,\ k=5)+2E_p \rightarrow H(n=1)+H(n=2) \ . \tag{3.4}$$

The atom of hydrogen $H(n=2)$ would then be ionized through the simultaneous absorption of two additional photons. The preceding remark on saturation applies to these two-photon transitions, so that the total probability of obtaining H^+ ions would be only governed by the five-photon process, which would be slow because of the change of rotational state.[20] Lastly, a direct dissociation of the molecule is possible through the simultaneous absorption of seven photons into the continuum of the excited $B'\,{}^1\Sigma_u$ or $C\,{}^1\Pi_u$ state. However the seven-photon process is incompatible with the observed slope $K=5.7$.

C. Formation of the Observed Molecular Hydrogen Ion $H_2{}^+$

A similar mechanism may be proposed to explain the observed slope $K=6.4\pm 0.5$ of H_2^+ for the multiphoton ionization of the hydrogen molecule. This multiphoton ionization may result either from a direct seven-photon transition to the $(v=4)1s\sigma_g$ molecular-ion state or through autoionization of higher Rydberg states of H_2. The $(3p\sigma)\,B'\,{}^1\Sigma_u$ represents one of the lower $np\sigma\,{}^1\Sigma$ and $np\pi\,{}^1\Pi$ Rydberg states. The potential curves of these higher Rydberg states converge towards the ground state $1s\sigma_g$ of H_2^+ as $n\rightarrow\infty$. These Rydberg states can be described by an electron outside a vibrationally excited molecular ion core. These states play a significant role in photoionization of H_2 because they are strongly coupled with the ionization continuum due to vibrationally induced autoionization through radiationless vibrational relaxation of the core. The rates of autoionization for specific vibrational-electronic states are very fast (10^{10}–10^{12} sec^{-1}). This gives rise to considerable broadening of some absorption lines of the H_2 spectrum near the ionization threshold.[21] The energy of seven photons is equal to the transition energies from the $(v=0)\,X\,{}^1\Sigma_g$ state of H_2 into the higher $(v=5)\,8p\pi$ and $(v=5)\,8p\sigma$ Rydberg states, or into the $(v=4)\ 1s\sigma_g$ state of H_2^+. Thus, the multiphoton ionization of H_2 may be due to various processes. For example, a direct seven-photon ionization may occur, that is,

$$H_2+7E_p \rightarrow H_2^+1s\sigma_g(v=4)+e \ . \tag{3.5}$$

Alternatively, a seven-photon absorption may occur so that higher Rydberg states are excited, that is,

$$H_2+7E_p \rightarrow H_2^*\ 8p\sigma(v=5) \text{ or } H_2^*\ 8p\pi(v=5) \tag{3.6}$$

followed by an autoionization of these higher Rydberg states to give $H_2^+\,1s\sigma_g\,(v=3)+e$ with a rate greater than 10^{10} sec^{-1}.[21] Finally, we may consider a six-photon excitation of the intermediate state, that is,

$$H_2+6E_p \rightarrow H_2\ E,\ F\,{}^1\Sigma_g^+\ (v=21,\ k=2) \tag{3.7}$$

followed by the absorption of one additional photon to give the $(v=5)\,8p\sigma$ or $(v=5)\,8p\pi$ Rydberg states and then autoionization in $H_2^+\,1s\sigma_g\ (v=3)$ or followed by the one-photon absorption to give $H_2^+\ 1s\sigma_g\ (v=4)$ $+e$.

The probability of the one-photon dissociation, that is,

$$H_2^+ + E_p \rightarrow H + H^+ \text{ at } \lambda = 0.53\ \mu \tag{3.8}$$

may be calculated using analytical expressions for photodissociation cross sections derived within the framework of the theory of multiphoton transitions in homopolar diatomic molecules.[22] The values $\sigma_3=7.9\times 10^{-27}$ cm^2 and $\sigma_4=4.3\times 10^{-23}$ cm^2 for the $(v=3,4)\,H_2^+$ levels give rise to the dissociation probability 8.4×10^4 sec^{-1} and 4.5×10^8 sec^{-1}, respectively, at $\Gamma=4\times 10^{12}$ W/cm^2. Thus the dissociation rate of the $(v=4)$ H_2^+ level would be high enough for the subsequent total dissociation of molecular hydrogen ions ($\tau\simeq 2$ nsec) during the laser pulse. On the other hand, the lifetime of the autoionization channel final state H_2^+ $(v=4)$ is equal to 10^{-5} sec. The experimental observation of molecular ions H_2^+ indicates that autoionization through high Rydberg states seems to be responsible for producing H_2^+. Furthermore, the respective atomic and molecular ion formation processes from the $(v=0)$ level of the H_2 ground state seem to be well separated.

IV. INTERACTION OF 0.53-μ LASER RADIATION WITH ATOMIC HYDROGEN

In a second set of experiments, the molecular-hydrogen beam was dissociated with rf power. The interaction between the 0.53-μ laser radiation and the hydrogen beam was investigated for three different dissociation fractions; 30, 50, and 80%. Figure 5 shows the variation of the resulting atomic hydrogen ions H^+ as a function of the laser intensity under these conditions. Compared to the number of H^+ ions formed from a molecular hydrogen beam, a significant increase in the number of H^+ ions is observed at a given laser intensity when the hydrogen beam contains 30% hydrogen atoms. This increase can be explained as follows. The population of the vibrational levels of the H_2 electronic ground state is described by the Boltzmann distribution. The H_2 molecules are heated when the rf power is turned on. The vibrational-level population does not diminish so rapidly as the number of vibrational level increases. As for the formation of H_2^+ ions, a seven-photon absorption takes place between the $(v\geq 2)$ levels of the ground

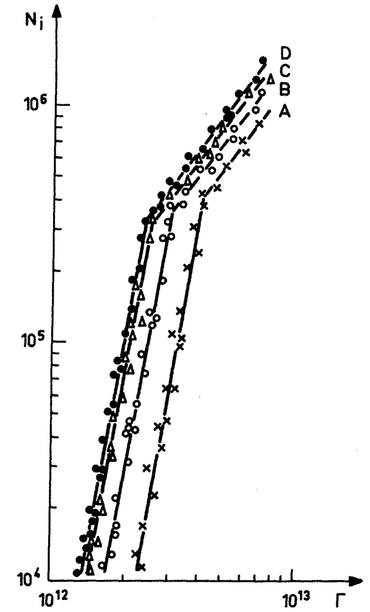

FIG. 5. Variation of the number of atomic ions H^+ formed as a function of the laser intensity Γ from: A, molecular hydrogen only; B, molecular hydrogen 30% dissociated by rf power; C, molecular hydrogen 50% dissociated; and D, molecular hydrogen 80% dissociated.

state of H_2 and the $(v \geqslant 8)$ levels of the $1s\sigma_g$ state of H_2^+ or with the dissociation continuum. Molecules in these levels dissociate rapidly into the $2p\sigma_u$ repulsive state through the absorption of one additional photon.[23] The same remark applies to a six-photon transition from vibrationally excited $(v \geqslant 2)$ levels of the H_2 ground state to the continuum of the E, $F\,{}^1\Sigma_g$ state. This dissociation of H_2^+ explains on the one hand the very fast decrease of the number of H_2^+ ions, and on the other hand the very significant increase of the number of H^+ ions when the rf power is turned on. This increase of the number of H^+ ions is much higher than the contribution due to multiphoton ionization of atomic hydrogen resulting from the preliminary dissociation of molecular hydrogen with rf power for a 30% dissociation. For high degree of rf power dissociation of molecular hydrogen, the dominant contribution to the formation of H^+ ions seems to result from multiphoton ionization of hydrogen atoms resulting from an 80% rf dissociation of molecular hydrogen. The observed slope $K = 5.8 \pm 0.5 \simeq K_0 = 6$ for the formation of H^+ seems to result from a direct six-photon ionization of atomic hydrogen without passing through any intermediate excited atomic levels. The six-photon ionization probability W of the atom of hydrogen can thus be determined. One can utilize the relationship $W\tau_6 = 1$ for the laser intensity corresponding to the saturation point in the number of H^+ ions. Thus, $W = 9 \times 10^{7\pm 0.6}\,\mathrm{sec}^{-1}$ for the laser intensity $\Gamma = (2.4 \pm 1.0) \times 10^{12}$ W/cm^2. On considering the curves A and D of Fig. 5, it should be pointed out that a very significant error in the determination of the multiphoton ionization probability of atomic hydrogen would have taken place if the molecular hydrogen were not first 80% dissociated by rf power.

V. COMPARISON BETWEEN THE CALCULATED AND THE MEASURED SIX-PHOTON IONIZATION PROBABILITY OF ATOMIC HYDROGEN

Figure 6 shows the variation of the experimental six-photon ionization probability for atomic hydrogen as a function of the laser intensity. This figure also shows the six-photon ionization probability of atomic hydrogen calculated at the wavelength $\lambda = 0.53\ \mu$ using perturbation theory by Gontier and Trahin,[10] Gold and Bebb,[7] and Morton.[8] A significant disagreement between experimental and calculated probabilities is observed, even if experimental errors are taken into account. The experimental probability is greater than the corresponding calculated probability. This apparent discrepancy may be explained in terms of coherence of the laser radiation. It should be pointed out that theoretical calculations assume the laser radiation to be single mode. Multiphoton-ionization probability W varies as

$$\overline{\Gamma^{K}} = \phi(K)(\overline{\Gamma})^{K} , \tag{5.1}$$

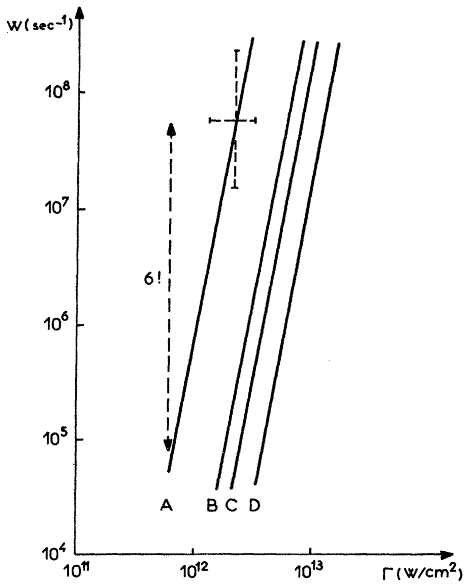

FIG. 6. Variation of the six-photon-ionization probability W of atomic hydrogen as a function of the laser intensity Γ. Comparison of the experimental probability A with the calculated probabilities B given by Gontier and Trahin (Ref. 10); C, Gold and Bebb (Ref. 7); and D, Morton (Ref. 8).

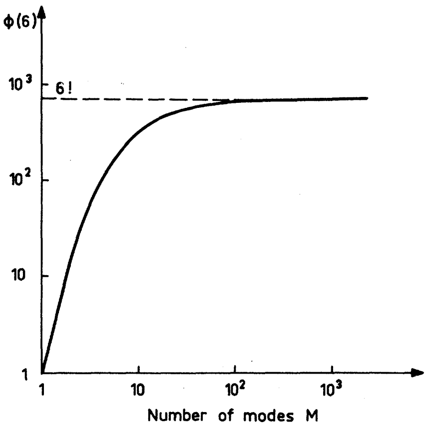

FIG. 7. Sixth-order normalized correlation function $\phi(6)$ vs the number of modes M of the laser field.

where $\phi(K)$ is a Kth-order correlation function.[24] $\phi(K)=1$ for a single-mode laser. However, the laser radiation used in the experiment reported here contains numerous independently oscillating modes. The spectrum of Nd-glass laser radiation generally contains approximately 10^3 axial modes, whereas the number of axial modes generated by a ruby laser is about ten. The influence of the quantum statistics of light on the multiphoton transition probability has been theoretically investigated in the last few years. Figure 7 shows the variation of the calculated six-order normalized correlation function versus the number of modes. The modes of oscillation are assumed to be stationary, to be statistically independent of one another, and to have intensities of comparable magnitude.[24] It can be seen that for laser fields involving few modes, the correlation function undergoes a rapid variation when the number of modes is changed. It should be pointed out that the light becomes incoherent as the number of modes becomes increasingly large. The limiting value of the sixth-order normalized correlation function is shown to be 6!. Thus, it would seem reasonable to assume that $\phi(K)=K!$ for Nd-glass laser radiation, and hence the measured multiphoton ionization probability would be $K!$ times the corresponding calculated probability. Thus, in Fig. 6 the measured six-photon-ionization probability of atomic hydrogen is found to be in good agreement with the calculated six-photon ionization probability when the factor 6! is taken into account. A similar result was obtained in the four-photon ionization of potassium atoms with Nd-glass laser radiation.[25] This agreement would demonstrate the validity of perturbation theory in laser field of some 10^{12} W/cm^2. It would not be the same in laser fields much higher than 10^{13} W/cm^2, where the usefulness of perturbation theory would be in doubt. However a theoretical nonperturbative method has been recently presented by Reiss.[26] This method is valid for arbitrarily high laser intensities and will prove useful for laser intensities of 10^{14}– 10^{15} W/cm^2 generated from mode-locked Nd-glass lasers.

VI. CONCLUSION

In performing the experiments described in the previous sections, the purpose was to investigate multiphoton dissociation and ionization processes of molecular hydrogen, and, primarily, to determine the multiphoton-ionization probability of atomic hydrogen resulting from a molecular hydrogen beam which was 80% dissociated with radiofrequency power. This determination was only found to be possible for a highly dissociated beam of hydrogen. In this condition six-photon ionization of atomic hydrogen was observed. This experimental six-photon-ionization probability proves to be higher than the corresponding calculated six-photon-ionization probability of atomic hydrogen in the ground state. However, the experiments have been carried out with the second harmonic generation of a Nd-glass laser for which the spectrum contains about 10^3 axial modes. It will be necessary to directly investigate the influence of photon statistics of the laser radiation on the multiphoton-ionization process with a laser of variable coherence. This experiment will test the validity of the correction term $K!$ which, when taken into account gives good agreement between theoretical and experimental values of the six-photon ionization probability of atomic hydrogen.

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[20] The rotational structure of the $0 \rightarrow 3$ band of the $X \rightarrow B$ system of H_2 is much more open than that of E, F ($v=21$) level. Successive rotational lines for $k=5$ are separated by about 150 cm^{-1}, which is the condition for resonance. Multiphoton absorption may be only satisfied by the single rotational $B\ \Sigma_u$ ($v=3$, $k=5$) level achievable through five-photon absorption for which the resonance gap lies close to the bandwidth of the laser radiation.

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Determination of the H-He Potential from Molecular-Beam Experiments

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Absolute total cross sections for D-He scattering were measured with an accuracy of 1% at 0.3, 0.5, and 0.6 eV. These new data were combined with published relative measurements of the velocity dependence of the H-He cross section in a χ^2 minimalization procedure. The resulting potential $V(R)$ as function of internuclear separation R is precisely determined in the repulsive range $1.7\ \text{Å} < R < R_0$ with $V(R_0) = 0$. Several multiparameter potential models were tried. Of these the four-parameter *ansatz* $V(R) = A(e^{-\alpha R} - e^{-\alpha R_0})$ (for $R < R_0$) $= 4\epsilon[(R_0/R)^{12} - (R_0/R)^6]$ (for $R > R_0$) was found to describe the data best. Parameters obtained are $A = 70.00$ eV, $\alpha = 3.383\ \text{Å}^{-1}$, $R_0 = 3.20$ Å, and $\epsilon = 0.39$ meV. The well depth ϵ, to which the measurements are insensitive, is chosen to match the theoretical van der Waals constant $C_6 = 2.83$ a.u. The potential is compared with results of *ab initio* calculations, and a probable ϵ range is discussed. For $0.35 \lesssim \epsilon \lesssim 0.70$ meV the calculated low-energy cross section shows a pronounced Ramsauer-Townsend minimum. The possibility of its observation is pointed out.

The H-He system is the simplest three-electron diatom and has been the object of numerous quantum-chemical *ab initio* calculations,[1-6] some of which have been performed over the entire (i.e., short, intermediate, and long) range of internuclear separations.[1,3,4,6] The most recent calculations show van der Waals wells,[3-6] the depth (ϵ) of which differs within an order of magnitude.

The H-He system is also of astrophysical interest. For instance, the infrared line shape in translational absorption is determined by the potential curve and has been suggested as being a possible factor in the opacity of late-type stars.[7] Furthermore, the potential is also needed to interpret hyperfine pressure-shift measurements on H in a He buffer gas.[3,6]

In order to get direct experimental information on the intermediate range of the potential, we have