Continuum-state selectivity in hydrogen in Stark fields by charge-shape tuning

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We present numerical calculations of the photoionization of atomic hydrogen in the presence of a strong dc electric field, using three-photon excitation with two-photon resonance with intermediate Stark states. The systematics of the dependence of the cross section on the intermediate Stark states is calculated for the $n=2$ to $n=9$ manifolds. Our results indicate that one can use "charge-shape tuning" to selectively excite and enhance Stark-induced giant dipoles near $E=0$ in hydrogen without the excitation of the overlapping continuum. Frequency selectivity can be used to excite from $1s$ (spherical charge) an intermediate whose charge is focused along the field followed by another excitation to the giant dipoles. Charge tunability can be achieved by controlling the degree of focusing by choice of the field and intermediates.

In the last few years there has been much interest in the effect of electric fields on the structure and interaction of atoms with other atoms near $E=0$.[1–18] Recently, we studied the effect of an external dc electric field imposed on atomic hydrogen and showed that it can be used to construct nearly one-dimensional atoms that spontaneously ionize.[17–18] These have electronic charge distributions that are highly extended along the field, and may have enormous dipole moments ("giant dipole" atoms). In the $E\geq 0$ region they spontaneously ionize in times on the order of 10^{-12} s (width of a few wave numbers). Moreover the excitation of a given giant dipole state from the ground state necessarily results in the simultaneous excitation of the smooth continuum; with a branching ratio favoring the smooth continuum; thus these giant dipoles appear as weak broad modulations embedded in the otherwise smooth underlying continuum (so-called "Stark-induced modulations").

Recently we showed theoretically and experimentally that the branching ratio for the excitation of the dipoles or the strength of the modulations can be enhanced using multiphoton excitation rather than a single photon excitation.[17,18] Our previous work showed that a two-step process via the $n=2$ and $n=3$ intermediate Stark manifolds instead of a one-step process enhances the strength from 20.4% to 60% and 89%, respectively (at 16.8 kV/cm).

In this brief report we study in more detail the theoretical aspects of the two-stage process. Our calculations indicate that the spectral distribution of any final channel in the $E\geq 0$ region is composed of a resonance corresponding to a field-induced giant dipole and a continuum that opens up at its peak and builds up exponentially thus giving a large degree of overlap with the giant dipoles. Our calculations indicate that selective excitation of the giant dipole resonances without the excitation of the continuum can be achieved by what we call "charge shape tuning." In this method one can tune the shape of the charge distribution of the intermediate state to match that of the resonance in the $E\geq 0$. Such tuning can be achieved by the appropriate choice of the quantum numbers of the intermediate state: n,n_1,n_2,m_l. The most selective tuning is achieved by a choice of $m_l=0$, $n_2=0,n_1=n-1$ with n as large as possible. The selective excitation is manifested by narrowing of the yield and enhancement of the peak cross sections and consequently in the increase in the visibility or strength of the Stark induced modulations.

The structure of the giant dipole atom induced by the dc electric field Fz in the $E\geq 0$ region was determined numerically. The technique we used was described in detail in an earlier paper.[17] The electric field used in all of the present studies is 16.8 kV/cm, which is the field we used in previous theoretical and experimental studies. Moreover, the polarization of the radiation in all of the studies is taken to be parallel to the dc field so that we have a final $m_l=0$ channel. The intermediate states of interest originate from the manifolds $n=2$–9. The low-lying wave functions are essentially unaffected by the electric field. The states originating from $n=5$–8 are slightly affected by the field (to 5–8 %). Therefore, the matrix elements may be computed using the zero-field analytical expressions for the initial states. The states we will be using as intermediates are those states for which we have $n_1=n-1$, the maximum possible n_1 parabolic quantum number. These components are the bluest components in each manifold and their charge distributions are extended along the field becoming more and more focused or oriented along the external field as n_1 increases. In this Brief Report we will only consider the properties of the cross section of the second step of the excitation, that is for the excitation from the intermediate state to the final state.

Figure 1 gives three numerical lineshapes corresponding to the excitation of the $(n_1=18,\ n_2=0,m_l=0)$ channel from the intermediate channels (0,0,0), (1,0,0), and (8,0,0). These three channels are the ground state, $n=1$, the bluest Stark component of $n=2$, and the bluest Stark component of $n=9$. The peaks of the cross sections are normalized to unity for easy comparison. First we observe that the blue wings are much more extended than the red wings. This gives asymmetric profiles with the

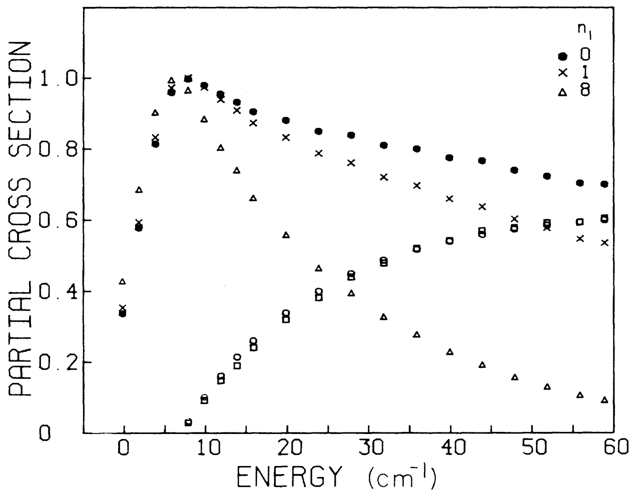

FIG. 1. Theoretical line shape of the transition between ($n_1=n-1$, $n_2=0$, $m_l=0$) and ($n_1=18$, $n_2=0$, $m_l=0$) for the cases $n=1$, 2, 9 in the presence of a dc electric field of 16.8 kV/cm and using laser radiation of polarization parallel to the external dc field. The peaks of the line shapes are normalized to unity. Also, the figure gives the difference of the blue wings for the case of $n=1$ and 9 (shown as □). We also show the prediction of an empirical expression for the difference given by $S=0.63(-\exp[-0.06(E-E_0)])$ (shown as ○).

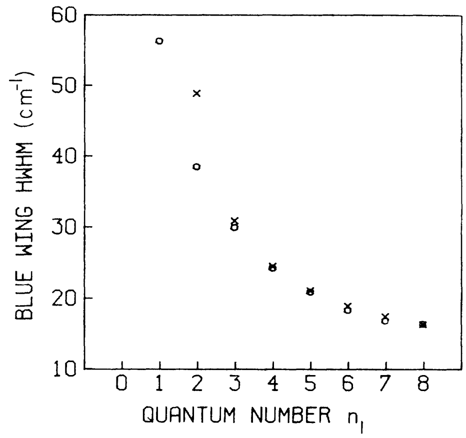

FIG. 3. The half width of the blue wing of the transition between ($n_1,n_2=0,m_l=0$) and ($n_1=18$, $n_2=0$, $m_l=0$) as a function of n_1 of the initial state in the presence of 16.8 kV/cm. ○ exact numerical calculation; × from the empirical expression $\Gamma=33.85/\ln n_1$.

red wings rising in about 10 cm^{-1} and the blue wings persisting over many tens of wave numbers. Secondly, where as we observe large variations in the blue wings, we find very little variation in the red wings as a function of n_1. The variation in the blue wing is such that a large degree of narrowing of the lines takes place as n_1 of the intermediate state increases.

Figure 2 shows the peak of the calculated cross section as a function of n_1. We see that the peak in the case of $n_1=8$ is about a factor of 300 larger than that of the ground state. Along with the results we give the cross sections given by the simple quadratic power law $\sigma=0.25n_1^2$. Apart from deviations at large n_1, where we see some saturation, we find it to be a reasonable approximation of the exact numerical values. Figure 3 gives the half width of the blue wing of the line as a function of n_1. We find that the width for $n_1\geq 2$ can be reasonably approximated by the simple formula $\Gamma=33.85/\ln n_1$. The values calculated from this formula are plotted in the same figure.

Finally we calculated the total cross section in the energy region of the 18,0,0 and 19,0,0 channels. Figure 4 gives the corresponding line shape in this region. The spectrum shows that we can attain spectra with very large

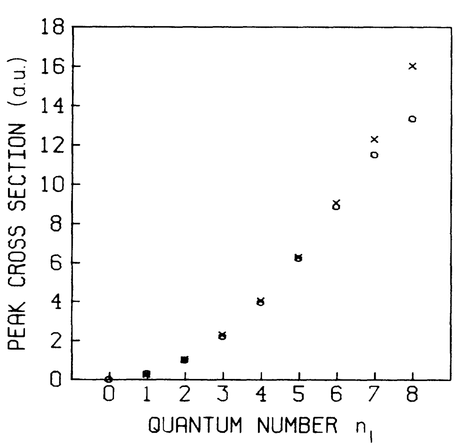

FIG. 2. The maximum cross section to the 18,0,0 state is plotted as a function of n_1 of the initial state n_1, $n_2=0$, m_l in the presence of 16.8 kV/cm and using radiation of π polarization. ○ represents exact numerical calculation; × from the empirical expression $\sigma=0.25/n_1^2$.

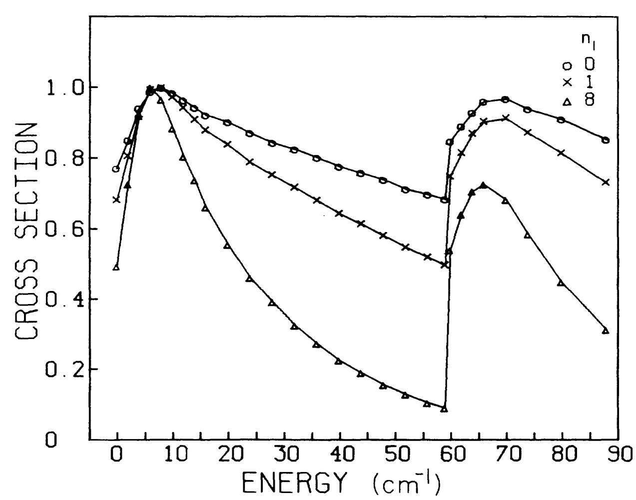

FIG. 4. Normalized total cross section as a function of energy near $E=0$ for excitation from $n_1=n-1=0$, 1, and 8 in the presence of a 16.8 kV/cm field and using radiation of π polarization.

visibility when the excitation is carried out from high n_1 states. From the total cross section we determined the strength of the "Stark-induced modulation" defined as $V=(S_{\max}-S_{\min})/(S_{\max}+S_{\min})/2$. Figure 5 shows these results as a function of n_1. We also plot in Fig. 5 the prediction of the empirical formula $V=200\,(1-e^{a(n_1+1)})$ for the strength of the resonance where $a=0.2$. This simple expression shows a linear rise for low n_1 values and saturation near high n_1.

In an attempt to analyze the origin of the blue wing of the line, we considered two line shapes: the line shape for excitation from the (8,0,0) state and the line shape for excitation from the ground state (0,0,0). We should note that our calculations show that the line shape changes very little when n_1 increases from 8 to 9, the highest state that is below the point $E=-2\sqrt{F}$. Thus the $n_1=8$ or $n_1=9$ represent the limiting line shape. This is also evident from Fig. 4 which gives the width of the blue wing as a function of n_1. Now we take the difference between the blue wings of these two line shapes (normalized ones), and show it along with the line shapes themselves in Fig. 1. The line shape of the difference indicates that it is due to an opening of a continuum channel. The channel build up completely at $E\sim 60\ \text{cm}^{-1}$, which is the location of the opening of next continuum channel associated with the state (19,0,0). Along with this difference we give in Fig. 1, the results of the empirical formula $S=S_0-S_8=S_c(1-e^{-(E-E_0)/E_c})$, where $S_c=0.63$, $E_0=7\ \text{cm}^{-1}$, the energy at which the threshold starts, and $E_c=16.6\ \text{cm}^{-1}$. The good agreement indeed indicates that the rise of the channel can be described by a saturation formula with the channel saturating or rising up completely to $\sim 60\ \text{cm}^{-1}$, which is the energy at which the next channel opens.

Thus we can, in general, consider the opening of a channel as an opening of a resonance that is associated with an oriented charge (giant dipole) and a continuum. The resonance can be isolated almost completely be exciting from a high n_1 state, and hence one can get its parameters from the (8,0,0) line shape. For example, from this line shape we find that, at the present field, the width at half maximum of the blue wing is $\simeq 16.6\ \text{cm}^{-1}$, whereas the width of its red wing is $\simeq 6\ \text{cm}^{-1}$.

We we will use charge distribution arguments to give an explanation of why the continuum channel is not excited when the excitation is carried out from a high n_1 initial state. First we note that the charge distribution of the oriented giant dipole state is tightly stretched or focused along the field with the orbit subtending a very small solid angle with respect to the nucleus. The continuum component, on the other hand, is not focused along the field, and hences subtends a good fraction of 4π with respect to the nucleus.

Now we describe the charge distribution of the initial Stark states. The ground state is spherically symmetric, hence its charge distribution subtends a solid angle of 4π with respect to the nucleus. The charge distribution of the Stark excited states are not spherically symmetric; they are somewhat stretched or focused along the field with the centers of their electronic charge shifted from the nucleus. The degree of stretching and shifting in the direction of the electric field is maximum for the bluest components of each manifold, $n_1=n-1$, and both effects become larger as n_1, or alteratively as n, increases. Alternatively we can describe this interm of solid angle considerations by saying that the charge distribution of the bluest component subtend a solid angle with respect to the nucleus that decreases as n_1 increases.

Thus by exciting from a variety of excited states that have a range of degree of focusing along the field and hence achieve what we call "charge-shape tuning" one can select to overlap with a variety of charge distributions of the final state. For example, we expect to have large overlap between the ground-state distribution (unfocused) and the continuum component (unfocused). As n_1 increases, this overlap is expected to weaken while simultaneously the overlap with the focused giant dipole resonance is expected to pick up strength. The pick up of strength of the giant dipole resonance was illustrated in Fig. 2.

We should emphasize that our introduction of the simple empirical formula in Figs. 1, 2, 3, and 5 does not constitute a fit of the numerical data nor is it an attempt to deduce analytical results. Such analytical dependence may be determined if one solves the problem semianalytically. This procedure is not the object of the present work. We should note, therefore, that we have no justification for these expressions at this time except that they fit reasonably good. However these kind of studies are interesting and plans for carrying them out are underway.

In conclusion, we have theoretically studied the two-stage excitation of electric field induced giant dipole atoms in the $E\geq 0$ instead of one-stage from the ground state of atomic hydrogen. The resonant intermediate states used of a given n manifold are Stark states whose charge distribution are extended most along the field (maximum value of n_1 possible). Our calculations indicate that the spectral distribution of any final channel is

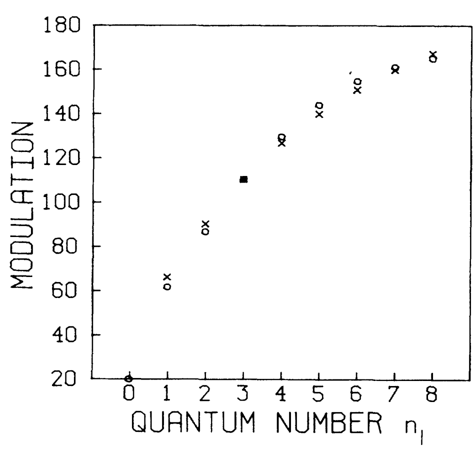

FIG. 5. The percentage of the Stark-induced modulation resulting from 18,0,0 (near $E=0$), as a function of n_1 of the initial state n_1, $n_2=0$, $m_l=0$ in the presence of 16.8 kV/cm and using radiation of π polarization. ○ is the exact numerical calculations; × the empirical expression $V=200(1-\exp[-0.2(n_1+1)])$.

composed of a resonance corresponding to a field-induced giant dipole, and a continuum that opens up at its peak and builds up exponentially thus giving a large degree of overlap with the giant dipole. Our calculations indicate that selective excitation of the giant dipole resonances without the excitation of the continuum can be achieved by what we call "change-shape tuning." In this method we tune the shape of the charge distribution of the intermediate state to match that of the resonances in the $E \geq 0$. Such tuning can be achieved by the appropriate choice of the quantum numbers of the initial state: n, n_1, n_2, m_l. The most selective tuning is achieved by a choice of $m_l = 0$, $n_2 = 0$, $n_1 = n - 1$ with n as large as possible. The selective excitation is manifested by narrowing of the yield, enhancement of the peak cross section and consequently in the increase in the visibility or strength of the Stark induced modulations.

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