Electric field ionization of highly excited sodium *nd* atoms

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Selective field ionization (SFI) is frequently used to detect atoms in high Rydberg states and to analyze Rydberg population distributions. In order to realize the full potential of this technique, however, a detailed understanding of the behavior of Rydberg atoms during passage from low to ionizing fields is required. The factors influencing this passage are discussed quantitatively and are illustrated by means of detailed SFI data obtained for Na 34*d* atoms.

I. INTRODUCTION

Atoms in high Rydberg states are frequently detected by use of selective field ionization (SFI) (Ref. 1) in which the atoms are ionized in an increasing electric field and the resulting electrons are detected as a function of applied field. This technique is attractive both because it provides unit detection efficiency and because it is suitable for atoms in states with large principal quantum numbers. In addition, because atoms in different Rydberg states evolve differently in an increasing field, it is in principle possible to infer the identity of Rydberg atoms from the field dependence of their ionization signal. However, a detailed, quantitative analysis of Rydberg-atom population distributions can only be undertaken using SFI if the response of the Rydberg atoms to the applied field is fully understood.

Previous studies[1–7] have provided a qualitative understanding of many aspects of the field-ionization process. In this work we have undertaken a more detailed study of the electric field ionization of Na 34*d* atoms considering quantitatively both the excitation of these atoms and their response to an increasing field. We have determined, at a selected ionizing-field slew rate, the separate SFI profiles appropriate to ionization of the $|m_l|=0$, 1, and 2 states that result from 34*d* excitation, thereby enabling synthesis of the SFI profile for any arbitrary mixture of these states. We have calculated the relative number of atoms in $|m_l|=0$, 1, and 2 states resulting from specific zero-field excitation conditions followed by either adiabatic or diabatic passage to intermediate fields. Synthesized SFI spectra obtained from the results of these calculations are found to be in excellent agreement with those observed experimentally. This agreement indicates that quantitative information concerning Rydberg-atom population distributions may be obtained from detailed analysis of SFI spectra.

II. EXPERIMENTAL CONSIDERATIONS

The apparatus is shown schematically in Fig. 1. Ground-state sodium atoms contained in a beam are excited to 34*d* states in a magnetically shielded region and in near-zero ($\lesssim 0.05$-V cm^{-1}) electric field by two-step laser-induced photoexcitation via the intermediate $3p\,^2P_{1/2}$ state. The excitation region is located between two parallel mesh grids across which the ionizing field is applied. The exciting lasers may be plane polarized either with their electric vectors parallel (π polarization) or perpendicular (σ polarization) to the direction of the ionizing field.

The excited atoms are ionized by a pulsed electric field produced by application of suitable potentials to the two mesh grids. The electrons liberated at ionization are detected by a Johnston electron multiplier whose output is fed to a time-domain multichannel analyzer (MCA). The MCA is started at the beginning of the ionizing voltage ramp and is stopped by the first electron pulse subsequently re-

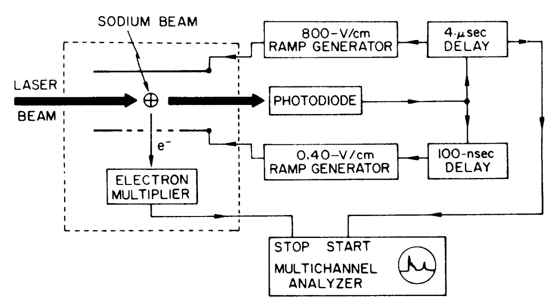

FIG. 1. Schematic diagram of the apparatus.

gistered by the detector. For sufficiently low count rates ($\lesssim 0.1$ per laser shot) the MCA stores a signal proportional to the probability of a field-ionization event per unit time as the ionizing field is increased. Measurement of the time dependence of the voltage applied to the grids then permits determination of the field strengths at which the ionization events occur and hence of the field-ionization signal per unit field increment.

III. RESULTS AND DISCUSSION

In order to analyze SFI data it is necessary to understand how the atoms respond as the applied field is increased from zero to the value at which ionization occurs. To facilitate this discussion we define four electric field regimes:

(1) Low. Stark energy shift much less than the fine-structure splitting.

(2) Intermediate. Stark energy shift greater than the fine-structure splitting.

(3) High. Excited state couples strongly with adjacent states within its own Stark manifold and, at higher fields, within different Stark manifolds.

(4) Ionizing. The limit of the high-field region where the atom spontaneously ionizes at a large rate ($\gtrsim 10^9$/sec).

In low field the states are best labeled by the quantum numbers n, l, j, m_j whereas at intermediate fields they are better described by the quantum numbers n, l, m_l, m_s. The component of the total angular momentum along the field axis is conserved and $m_j = m_l + m_s$. Since m_l remains a good quantum number during passage from intermediate to ionizing fields, it is therefore the low-to-intermediate field transition that determines the relative populations in each $|m_l|$ level that are crucial in determining the form of the SFI profiles.

Passage from low to intermediate fields may be discussed by reference to Fig. 2. This figure is constructed by energy ordering the states in both low and intermediate fields and then connecting each initial $|m_j|$ state to the final $|m_l|$ states into which it may evolve. If the electric field is increased sufficiently slowly there will be a gradual adiabatic evolution of each initial low-field state into a single corresponding intermediate-field state as shown in Fig. 2(a). If, however, the electric field is applied sufficiently rapidly the zero-field states will be projected diabatically onto the $|n,l,m_l,m_s\rangle$ states from which they are composed as shown in Fig. 2(b). As is evident from Fig. 2 the contribution from each of the initial $j = \frac{3}{2}, \frac{5}{2}$ states to the final $|m_l| = 0$, 1, 2

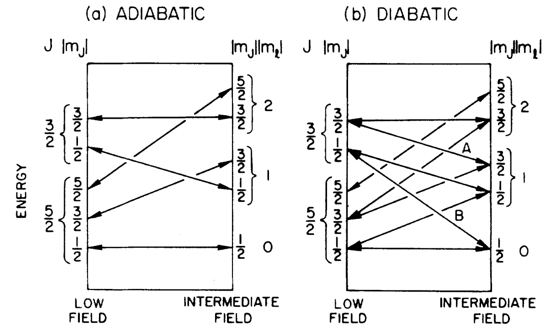

FIG. 2. Evolution from low to intermediate fields. The diagram for adiabatic passage is the same as given by Gallagher *et al.* (Ref. 1).

populations is strongly dependent on whether the passage from low to intermediate fields is adiabatic or diabatic.

The calculated relative productions of $|m_j| = \frac{1}{2}$ and $\frac{3}{2}$ states following photoexcitation to $nd\,^2D_{3/2}$ states via the intermediate $3p\,^2P_{1/2}$ state are presented in Table I for both π and σ laser polarizations. The behavior of these states during passage to intermediate fields will be illustrated using the states $|j = \frac{3}{2}, |m_j| = \frac{1}{2}\rangle$ as a specific example. If the passage is adiabatic, atoms in these states will evolve into states with only $|m_l| = 1$ [see Fig. 2(a)]. In the case of diabatic passage, atoms in $|j = \frac{3}{2}, |m_j| = \frac{1}{2}\rangle$ states will evolve into both $|m_l| = 0$ and 1 states [see Fig. 2(b)]. The relative populations in each $|m_l|$ state are given by the squares of the Clebsch-Gordan coefficients in the expansion of the initial $|j,m_j\rangle$ state in terms of product $|l,m_l\rangle|s,m_s\rangle$ states. For $|j = \frac{3}{2}, |m_j| = \frac{1}{2}\rangle$ states this expansion is

$$|\tfrac{3}{2}, \pm\tfrac{1}{2}\rangle = \mp(\tfrac{2}{5})^{1/2}|2,0\rangle \cdot |\tfrac{1}{2}, \pm\tfrac{1}{2}\rangle \pm (\tfrac{3}{5})^{1/2}|2,\pm 1\rangle\,|\tfrac{1}{2}, \mp\tfrac{1}{2}\rangle \qquad (1)$$

and thus 40% of the laser-excited atoms will yield $|m_l| = 0$ states, and 60% will yield $|m_l| = 1$ states following diabatic passage to intermediate fields. The calculated relative abundances of atoms in $|m_l| = 0$, 1, and 2 states for π and σ laser polarizations coupled with either diabatic or adiabatic passage to intermediate fields are given in Table I.

The transition from low to intermediate fields is accomplished by application of a small voltage step to the lower mesh grid. This step is applied ~ 100 nsec after laser excitation and provides an electric field that increases linearly to 0.4 V cm^{-1} with a slew rate that is adjustable over the range 0.1 to 50

TABLE I. Calculated relative production of $34d\ ^2D_{3/2}\ |m_j| = \frac{1}{2}$ and $\frac{3}{2}$ states for excitation via the intermediate $3^2P_{1/2}$ state, and the relative production of $|m_l| = 0$, 1, and 2 states following adiabatic and diabatic passage to intermediate fields.

Laser polarization	Low-field production $\lvert m_j \rvert$		Passage to intermediate fields: Adiabatic $\lvert m_l \rvert$			Diabatic $\lvert m_l \rvert$		
	$\frac{1}{2}$	$\frac{3}{2}$	0	1	2	0	1	2
π	100%	0	0	100%	0	40%	60%	0
σ	25%	75%	0	25%	75%	10%	30%	60%

$\mathrm{V\,cm^{-1}\,\mu sec^{-1}}$.

The subsequent transition to ionizing-field strengths is achieved by application of a high-voltage pulse to the upper mesh grid at a fixed time ($t \sim 4$ μsec) after laser excitation. The characteristics of this pulse are such that the electric field in the excitation region increases to $\sim 800\ \mathrm{V\,cm^{-1}}$ at a slew rate of $\sim 10^9\ \mathrm{V\,cm^{-1}\,sec^{-1}}$. The response of an atom to this time-dependent field is strongly influenced by interactions between states of the same $|m_l|$ that lead to avoided crossings whenever states of the same $|m_l|$ in different Stark manifolds approach one another.[8,9] The ionization behavior of an atom then depends on whether these avoided crossings are traversed adiabatically or diabatically and this in turn depends on the slew rate of the ionizing field. For $n \sim 34$ and a slew rate of $\sim 10^9\ \mathrm{V\,cm^{-1}\,sec^{-1}}$ atoms in states with $|m_l| = 0$, 1 pass to ionization along predominantly adiabatic paths, while those in states with $|m_l| = 2$ typically pass along predominantly diabatic paths.[3]

The transition from low to intermediate fields is investigated by varying the rise time of the low-voltage step, while keeping the high-voltage pulse unchanged. In Fig. 3 are shown the ratios of the signals resulting from ionization of $34d$ $|m_l| = 2$ states, and from ionization of $34d$ $|m_l| = 0$ states, to the total ionization signal as a function of slew rate from low to intermediate fields. The $34\,^2D_{3/2}$ states are excited via the $3\,^2P_{1/2}$ state with σ polarization. At slew rates above 20 $\mathrm{V\,cm^{-1}\,\mu sec^{-1}}$ the ratios are essentially slew-rate independent, indicating that for these conditions all atoms pass diabatically from zero to intermediate fields. At slew rates below 0.50 $\mathrm{V\,cm^{-1}\,\mu sec^{-1}}$ the ratios are again slew-rate independent indicating that all atoms pass adiabatically from zero to intermediate fields. At slew rates between these values atoms will pass neither totally adiabatically nor totally diabatically from low to intermediate fields. Data pertaining to adiabatic passage from low to intermediate fields are therefore obtained at a slew rate of 0.31 $\mathrm{V\,cm^{-1}\,\mu sec^{-1}}$, and data pertaining to diabatic passage are obtained at a slew rate of 33 $\mathrm{V\,cm^{-1}\,\mu sec^{-1}}$. The $34d$ state is chosen for study because the slew rates required to produce both adiabatic and diabatic passage to intermediate fields can be readily obtained and because, as will become

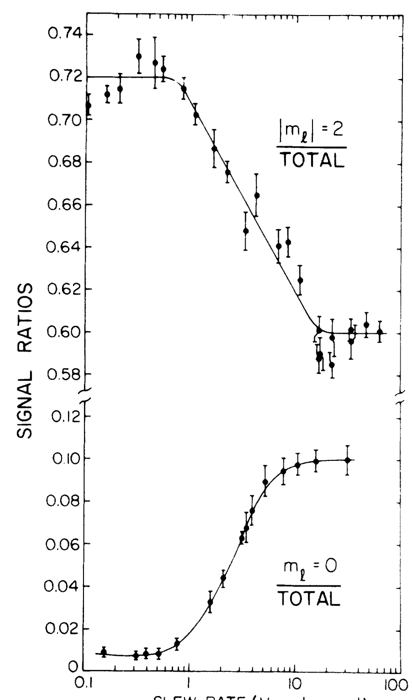

FIG. 3. Ratio of the signal resulting from ionization of $|m_l| = 2$ states (upper curve), and ionization of $|m_l| = 0$ states (lower curve) to the total ionization signal as a function of the slew rate from low to intermediate fields. Excitation of $34^2D_{3/2}$ via $3^2P_{1/2}$ with σ polarization.

evident, the $m_l=0$ and $|m_l|=1$ ionization features can be reasonably well separated.

With an appropriate choice of intermediate $3p$ state (i.e., $3^2P_{1/2}$ or $3^2P_{3/2}$), laser polarization, and zero-to-intermediate-field slew rates it is possible to produce atoms in either $m_l=0$ or $|m_l|=1$ states and ionization of these states can be studied separately. In order to obtain the SFI profile for $m_l=0$ states it is necessary to excite via the intermediate $3^2P_{3/2}$ state. Calculations similar to those previously discussed indicate that excitation with π laser polarizations, followed by adiabatic passage to intermediate fields, results in the production of excited atoms 98% of which are in $m_l=0$ states. The resultant SFI profile is shown in Fig. 4(a).

As evident from Table I, excitation via the intermediate $3^2P_{1/2}$ state with π laser polarizations, followed by adiabatic passage to intermediate fields, yields only $|m_l|=1$ states and their SFI profile is shown in Fig. 4(b). For all other excitation conditions more than one value of $|m_l|$ is produced. The locations of the $|m_l|=0$ and 1 ionization features on the electric field axis indicate that atoms in these states pass from intermediate to high fields along predominantly adiabatic paths.

Excitation with σ laser polarizations, followed by adiabatic passage to intermediate fields, yields the SFI profile shown in Fig. 4(c). Inspection of Table I shows that this profile corresponds to that for ionization of atoms, 25% of which have $|m_l|=1$ and 75% of which have $|m_l|=2$. Since the SFI profile for $|m_l|=1$ states is known, the field-ionization profile appropriate to $|m_l|=2$ atoms can then be obtained by subtracting the known contribution due to $|m_l|=1$ states. The resultant profile for $|m_l|=2$ states is shown, after renormalization, in Fig. 5(a). The feature at ~ 500 V cm^{-1} results from atoms that pass from intermediate to high fields along predominantly diabatic paths. However, a significant number of atoms in

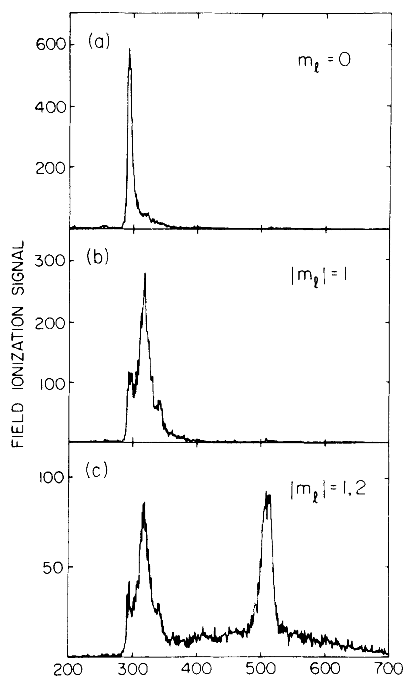

FIG. 4. SFI data obtained using an intermediate- to high-field slew rate of $\sim 1\times 10^9$ V cm^{-1} sec^{-1}. The data are normalized to equal initial Rydberg populations. (a) SFI profile for the $m_l=0$ state (excitation via $3^2P_{3/2}$ state with π laser polarization and adiabatic passage to intermediate fields). (b) SFI profile for the $|m_l|=1$ state (excitation via $3^2P_{1/2}$ state with π laser polarizations and adiabatic passage to intermediate fields). (c) SFI profile for a mixture of $|m_l|=1,2$ states (excitation via $3^2P_{1/2}$ state with σ laser polarizations and adiabatic passage to intermediate fields).

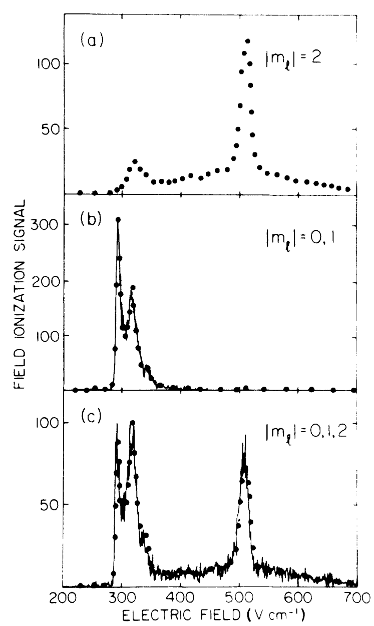

FIG. 5. Experimental (solid line) and synthetic (dotted) SFI profiles obtained using an intermediate- to high-field slew rate of $\sim 1\times 10^9$ V cm^{-1} sec^{-1}. The data are normalized to equal initial Rydberg populations. (a) SFI profiles for the $|m_l|=2$ state obtained from the data in Fig. 4 and Table I. (b) Synthetic and observed spectra for the mixture of $|m_l|=0$ and 1 states resulting from excitation via the $3^2P_{1/2}$ state with π laser polarizations and diabatic passage to intermediate fields. (c) Synthetic and observed SFI spectra for the mixture of $|m_l|=0$, 1, and 2 states resulting from excitation via the $3^2P_{1/2}$ state with σ laser polarizations and diabatic passage to intermediate fields.

$|m_l|=2$ states ionize at field strengths that correspond to predominantly adiabatic passage from intermediate to high fields.

Given the SFI profiles appropriate to atoms in $|m_l|=0$, 1, and 2 states it is then possible to synthesize SFI profiles appropriate to ionization of a mixture of atoms with any distribution of these $|m_l|$ values. Thus, as a test of internal self-consistency, the SFI profiles expected for diabatic passage to intermediate fields were determined using the data in Table I and a comparison between these and the corresponding experimental data is shown in Figs. 5(b) and 5(c). The agreement between the experimental and synthesized profiles is excellent, indeed the best fits between the synthesized and experimental profiles were obtained using precisely the values given in Table I.

The SFI profiles were found to depend on the time interval between laser excitation and the low-to-intermediate-field transition. Data showing the ratio of the signal resulting from ionization of $|m_l|=2$ states to the total ionization signal as a function of this time interval are presented in Fig. 6. The time dependence of this ratio is attributed to the presence of small off-axis residual electric and/or magnetic fields in the excitation region. The angular momentum $\vec{J}$ will precess in these fields resulting in changes in the projection of $\vec{J}$ on the direction of the ionizing field, thereby leading to changes in m_j (and hence m_l). This interpretation is reinforced by the fact that intentional application of small off-axis electric and magnetic fields in the excitation region resulted in changes in the time dependence of the SFI data. To avoid complications due to this effect the SFI profiles in Figs. 4 and 5 were recorded with the transition from zero to intermediate fields at the earliest possible time ($\sim 100-450$ nsec after laser excitation). Clearly, however, the possibility of effects due to the presence of small, off-axis electric and magnetic fields in the excitation region must be considered when analyzing SFI data. Indeed, measurements of the time evolution of SFI profiles may provide a useful technique with which to measure very small fields.

In order to emphasize the importance of the value of the intermediate-to-ionizing-field slew rate in determining SFI profiles, data were taken under the same conditions as shown in Figs. 4 and 5 except that the intermediate-to-ionizing-field slew rate was reduced by a factor of 5. The resulting SFI spectra are shown in Fig. 7. At the lower slew rates the $m_l=0$ and $|m_l|=1$ ionization features become completely resolved and states with $|m_l|=2$ ionize at lower field strengths than before. Indeed, as indicated by the absence of a diabatic peak at ~ 500 V cm^{-1}, few atoms in $|m_l|=2$ states now pass to

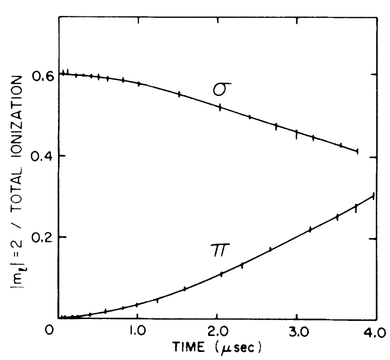

FIG. 6. Ratio of the signal resulting from ionization of $|m_l|=2$ states to the total ionization signal as a function of the time interval between excitation and the transition from zero to intermediate field. Excitation is via the $3^2P_{1/2}$ state with subsequent diabatic passage from zero to intermediate fields. Upper curve is for σ polarization and lower curve is for π polarization.

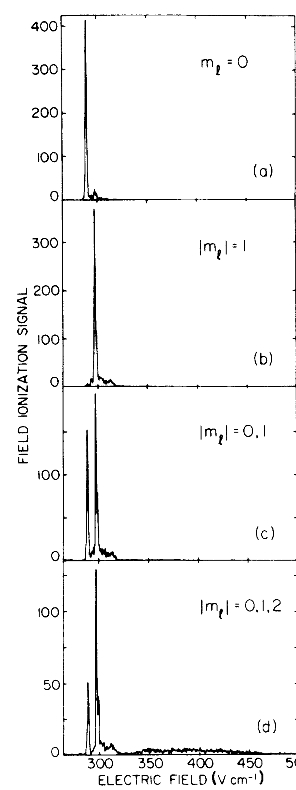

FIG. 7. SFI data obtained at an intermediate- to ionizing-field slew rate of $\sim 2\times10^8$ V cm^{-1} sec^{-1}. The excitation and passage to intermediate field conditions are (a) as in Fig. 4(a), (b) as in Fig. 4(b), (c) as in Fig. 5(b), (d) as in Fig. 5(c).

ionization along predominantly diabatic paths.

The internal self-consistency between the synthesized and observed SFI profiles indicates that quantitative analysis of SFI data is often possible provided proper account is taken of how the atoms respond to the increasing field. The SFI spectra depend on several parameters: (1) the $|j,m_j\rangle$ states initially excited which can be determined if the laser polarizations, intermediate $3p$ state, etc., are known; (2) the slew rate from low to intermediate fields which governs the distribution among the possible $|m_l|$ values; (3) the slew rate from intermediate to ionizing fields which determines the SFI profile appropriate to each individual $|m_l|$ state.

ACKNOWLEDGMENTS

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