Resolution of $|m_l|$ and $|m_j|$ Levels in the Electric Field Ionization of Highly Excited d States of Na†

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In the electric field ionization of highly excited s, p, and d states of Na, we have observed one, two, and three ionization thresholds, respectively, reflecting the number of high-field $|m_l|$ states coming from the s, p, and d states. Our observations indicate that the passage from low to intermediate field is adiabatic, and as a consequence we are able to achieve selective ionization of essentially degenerate low-field $|m_j|$ fine-structure levels.

The basic features of the electric field ionization of the lower excited states of atoms, in particular hydrogen, were investigated theoretically and experimentally soon after the development of quantum mechanics.[1] However, the extensive investigations of the higher-lying levels was to come much later. For example, only relatively recently Bailey, Hiskes, and Riviere[2] have done extensive numerical calculations of electric field ionization rates for many Stark states of H of n up to 20 using the method of Rice and Good.[3] In their experiments with fast hydrogen beams Il'in[4] and Bayfield and Koch[5] have investigated the ionization properties of hydrogen from $n = 9-23$ and $n = 63-69$, respectively. These experiments have verified the general n dependence of ionization predicted by Bailey, Hiskes, and Riviere.

Detailed studies of the ionization of single n, l states has only become possible with the advent of the tunable laser. Using laser techniques both Stebbings *et al.*,[6] studying Xe f states, and Ducas *et al.*,[7] studying Na s and d states, demonstrated that the threshold electric field for ionization is sharply defined and varies as n^{*-4}, where n^* is the effective principal quantum number. The thresholds are expected to be sharp because the field ionization rate increases exponentially with the strength of the ionizing electric field. The widths of the thresholds observed for the Xe f states and the Na d states are considerably wider than the Na s states, presumably because of the presence of more than one m_l state in the higher L states. We follow the usual convention that L, S, and J are the electron's orbital, spin, and total angular momenta and that m_l, m_s, and m_j are their projections in the field direction. The effects of nuclear spin are unimportant.

Here we report the resolution of the ionization thresholds of the $|m_l|$ states in the field ionization of highly excited p and d states of Na. Our observations indicate that at least the passage from zero field to intermediate field is adiabatic, allowing us to observe selectively the ionization of each of the low-field $|m_j|$ fine-structure levels.

Our experimental approach is quite similar to that used by Ducas *et al.*[7] An atomic beam of sodium at a density of 10^9 cm^{-3} passes between two electric field plate 1.11 cm apart where it is excited by two pulsed dye lasers. The dye lasers are tuned to the $3s$-$3p$ and $3p$-$3ns$, np, or nd transitions at ~5900 and ~4100 Å, respectively [we apply a low (~30 V/cm) field to allow the pumping of the $3p$-np transition]. Typically, the lasers have a pulse of 4 ns, pulse energy of 25 μJ, and a linewidth of 0.15 Å. A positive high-voltage pulse is applied to one of the electric field plates 0.2 μs after the laser pulses, field ionizing the highly excited atoms.

The ionizing pulse has a rise time of ~0.3 μs and stays within 5% of the peak voltage for 0.2 μs before decaying. Thus, what we observe as a threshold field for ionization corresponds to a field high enough that the ionization rate is 10^7 s^{-1}. The ions formed are accelerated toward the other electric field plate, a grounded grid, and pass through it onto a Venetian-blind electron multiplier. The multiplier is wired for pulsed operation to provide a linear response over a wide dynamic range.

The most important feature of our observations is the resolution of the $|m_l|$ states in the field ionization of highly excited Na. For s, p, and d states we saw, respectively, one, two, and three thresholds for ionization, reflecting the number of $|m_l|$ states for each of the s, p, and d states. Here we shall confine our attention to the ionization of the d states. In general we saw three thresholds in the ionization of the $d_{5/2}$ state and only two in the ionization of the $d_{3/2}$ state. A

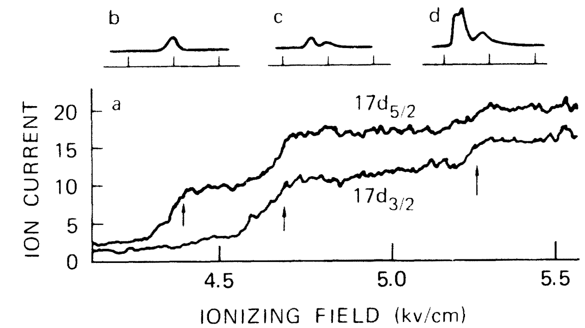

FIG. 1. (a) Experimental traces of the ion current versus peak ionization voltage for the $17d_{3/2}$ and $17d_{5/2}$ states. The approximate locations of the $|m_l| = 0$, 1, and 2 thresholds are indicated by arrows. (b), (c), (d) Oscilloscope traces of ion signals at different peak ionizing fields. In each case the center time marker corresponds to the peak of the ionizing high-voltage pulse. The horizontal scale is 200 ns/div. (b) $|m_l| = 0$ ion pulse, peak field = 4.58 kV/cm. (c) $|m_l| = 0$ followed by $|m_l| = 1$ ion pulse, peak field = 4.98 kV/cm. (d) $|m_l| = 0$ then overlapping $|m_l| = 1$ followed by $|m_l| = 2$ ion pulse, peak field = 5.27 kV/cm.

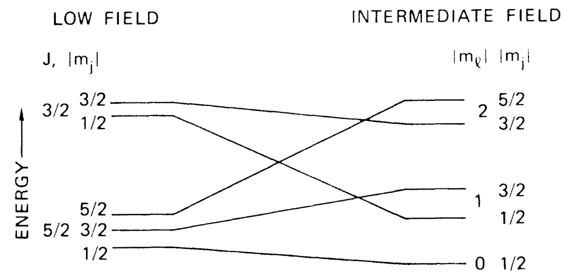

FIG. 2. Correlation diagram between low and intermediate field showing the connection between the low-field $|m_j|$ states and the intermediate-field $|m_l|$ states.

typical example of this is shown in Fig. 1(a), which shows traces of the ionization currents for both the $17d_{3/2}$ and $17d_{5/2}$ states as a function of peak ionizing field.

Since the field which we use to ionize the atoms increases in time from zero to a peak value, the field resolution of the steps shown in Fig. 1(a) is reflected in the time resolution of the ion peaks shown in Figs. 1(b), 1(c), and 1(d). The time-resolved ion signals for the ionization of the $17d_{5/2}$ state are shown for several peak ionizing field values. Figure 1(b), taken at a peak field of 4.58 kV/cm, shows the ion peak from the first state to ionize only. Figure 1(c), at 4.98 kV/cm, shows two well-resolved peaks. Note that the first peak appears earlier in time than in Fig. 1(b) because its threshold field is reached earlier in the ionizing pulse. Figure 1(d), at 5.27 kV/cm, shows all three $|m_l|$ peaks. The first two peaks appear even earlier than in Fig. 1(c), at a time when the ionizing field is increasing rapidly. Since the ionization rate increases exponentially with the field, the ionization rate for each of these states increases to a rate far above the threshold rate in a time $\ll 10^{-7}$ s, and as a result the observed peaks are narrower. The rapid increase of the field also reduces the time interval between reaching the first two thresholds with the result that we are unable to resolve completely the resulting two ion peaks. The clear time resolution of the three peaks is a distinct asset since we can selectively observe ions from only one $|m_l|$ even when all three $|m_l|$ states are ionized.

We have assigned the $|m_l|$ values to the ionization thresholds by establishing the connection between the low-field state to which the atom is excited and the high-field state from which it is ionized and by performing some simple polarization experiments with the laser to selectively populate $|m_j|$ fine structure states.

To characterize the passage from low to high fields, it is convenient to divide the electric field into three regimes; low field, where the spin-orbit interaction is stronger than the Stark effect; intermediate field, where the Stark effect is stronger than the spin-orbit interaction but not strong enough that the Stark manifolds from adjacent n states overlap; and high field, where the Stark manifolds are crossed. For the $17d$ state, low field < 10 V/cm < intermediate field < 1000 V/cm < high field. The observation that we see at most three thresholds tells us that each state follows a unique path from low to high field. Furthermore, the branching of the $d_{3/2}$ state into only two implies that the passage from low-field $|m_j|$ states to intermediate-field $|m_l|$ states is adiabatic with respect to $|m_j|$. Thus, we can connect the low- and intermediate-field levels by applying the no-crossing rule, i.e., states of the same m_j do not cross in passing from low to intermediate field. The low-field states are familiar fine-structure levels, $J=\frac{3}{2}$ and $J=\frac{5}{2}$, which are inverted ($J=\frac{3}{2}$ lies above $J=\frac{5}{2}$).[8] To determine the ordering of the intermediate-field $|m_l|$ states, we constructed the correlation diagram of Fig. 2 by applying the following criteria: inversion of the fine structure levels, adiabatic passage from low to intermediate field (no $|m_j|$ crossings), and the branching of the $J=\frac{5}{2}$ state to three $|m_l|$ levels and the $J=\frac{3}{2}$ state to two (our

TABLE I. Threshold fields for ionization in kV/cm (the field which produces an ionization rate of 10^7 s^{-1}). The experimental values are ± 4%.

		Na d state, experiment				
n	H_{theory}[a]	$\lvert m_l \rvert = 0$	$\lvert m_l \rvert = 1$	$\lvert m_l \rvert = 2$		
15	10.0	7.42	7.74	8.62		
16	8.4	5.48	6.05	6.43		
17	6.6	4.36	4.63	5.17		
18	5.2	3.40	3.53	3.88	4.11	4.32
19	4.2	2.74	2.81	3.03	3.28	3.54
20	3.4	2.22	2.30	2.48	2.62	2.73

[a]See Ref. 2.

experimental observation). Figure 2 is unique; no other ordering of $|m_l|$ states satisfies all three criteria. Figure 2 shows clearly the connections between the low- and intermediate-field states. Equally important, it identifies $|m_l| = 0$ as the state which comes only from the $J = \frac{5}{2}$ fine-structure level. Consequently, the first threshold in the ionization of the $d_{5/2}$ state is due to $|m_l| = 0$. Finally, it is worth stressing that it is the adiabatic nature of the low- to intermediate-field passage which enables us to selectively ionize the low-field $|m_j|$ fine-structure levels.

To assign the $|m_l| = 1$ and 2 states, we applied a low (~ 3 V/cm) dc field during the laser pulse. When the laser pumping the $3p$-nd transition was polarized so that $\vec{E}_{\text{laser}} \parallel \vec{E}_{\text{dc}}$, the third ionization peak disappeared. When the laser was polarized so that $\vec{E}_{\text{laser}} \perp \vec{E}_{\text{dc}}$, the third peak was stronger relative to the others. This clearly identifies the third threshold for ionization as $|m_l| = 2$ and the second therefore as $|m_l| = 1$.

In Table I we list the observed $|m_l|$ ionization thresholds for the Na d states from $n = 15$ to 20 and the thresholds for the analogous H state, the lowest Stark state of an n manifold, calculated by Bailey, Hiskes, and Riviere[2] assuming diabatic passage from low to high field. The discrepancy between the experimental Na values and theoretical H values simply indicates that in Na the passage is not completely diabatic. In their mapping of the $|m_l| = 0$ and 1 Stark levels of the $n = 15$–16 levels of Na, Littman *et al.*[9] showed very clearly that in high field states of the same $|m_l|$ repel strongly, because of the coupling between n states. This suggests that $|m_l| = 0$ and 1 states probably pass adiabatically from intermediate to high field. For $n < 18$ this is probably also true for $|m_l| = 2$. We attribute the observation of several $|m_l| = 2$ peaks for $n \geq 18$ to the partially diabatic passage of atoms in $|m_l| = 2$ states from intermediate to high field. As a result atoms do not follow a unique path from intermediate to high field and are ionized from different high-field states. Since these high-field states have different ionization thresholds, we observe multiple $|m_l| = 2$ ionization thresholds. Experiments are currently underway to investigate the passage from intermediate to high field in more detail. The ability to resolve the $|m_l|$ and $|m_j|$ substrates by their ionization thresholds expands the utility of the field ionization technique. For example, radio-frequency transitions between fine-structure states can be detected by the selective ionization of low-field $|m_j|$ states. We have already measured the polarizability and fine-structure intervals of several Na p and d states using this method which will be reported in a more detailed paper. We have previously pointed out that field ionization should be useful for laser isotope separation.[10] This selectivity reported here may lead to significant refinements in laser isotope separation.

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