Ion formation in laser-irradiated sodium vapor

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(Received 7 February 1983)

The formation of Na^+ and Na_2^+ in laser-irradiated sodium vapor has been studied using a nitrogen-laser-pumped dye laser. The effects of irradiation by this pulsed laser are substantially different from those reported previously using a cw device, but otherwise identical experimental conditions. In contrast to the earlier experiments, in which ions were observed only when the cw laser was tuned to a D line, relatively large "background" ion signals are observed at all excitation wavelengths, a result of multiphoton processes involving the Na_2 component of the vapor. It is shown that the laser bandwidth and pulse duration are extremely important parameters; a wide bandwidth effectively magnifying the importance of the dimer component of the vapor and a short pulse duration minimizing the effects of interactions between excited atoms and molecules.

I. INTRODUCTION

During the past few years there has been a great deal of interest in the production of plasmas and electrical discharges by laser irradiation of vapors, especially sodium. Lucatorto and McIlrath[1] have shown that irradiation of a rather dense column of Na vapor ($\sim 10^{16}$ cm^{-3}) with a dye laser tuned to a D line can cause complete ionization of the vapor. In addition to the intrinsic interest of this phenomenon the laser-produced plasma has been exploited as a new tool for absorption spectroscopy.[2] Stwalley and his co-workers have also produced plasmas by laser irradiation.[3] However, in their experiments the laser wavelength was not restricted to the D line; several wavelengths in the yellow region of the spectrum were employed. Since some of these wavelengths do not correspond to atomic resonances it is clear that excitation of the dimer component of the vapor, Na_2, plays a role.

Hertel and co-workers[4] have used laser-irradiated sodium vapor to produce infrared (ir) laser emissions. In this work yellow light from a pulsed dye laser was used to activate the vapor to form the required population inversion. While ir laser action was achieved when the excitation laser was tuned to a D line, it was also achieved when the excitation laser was tuned to optical wavelengths that do not coincide with *atomic* resonances. This again indicates participation by the neutral-dimer component of the vapor.

Because of the interest in laser-produced phenomena of this type several experiments have been performed that are directed toward study of the microscopic processes[5-21] that occur in laser-irradiated sodium vapor. These microscopic processes dictate the collective behavior of the plasma or discharge and are therefore important for a complete understanding of the phenomena. Among the interactions that contribute to this complex environment are those involving photons directly, such as $\hbar\omega + Na$ and $\hbar\omega + Na_2$, and indirectly, for example Na $3p$-Na $3p$ collisions.

The experiments that have been designed to investigate these microscopic processes have been performed under a variety of conditions. Vapor densities have been varied from $\sim 10^9$ cm^{-3}, characteristic of atomic beams,[12] to

$\sim 10^{14}$–10^{15} cm^{-3} in cell experiments.[7,10] Of course, the relative concentrations of the dimer ions differ as well since that concentration is temperature dependent. The lasers that have been employed have included both cw and pulsed; because of the characteristics of different pulsed lasers the pulse durations have ranged from ~ 10 nsec to greater than 1 μsec. Laser bandwidths have varied from those associated with multimode lasers to the narrow values achievable with single-frequency devices. Laser power densities have ranged from the low values associated with cw lasers (~ 1 W/cm^2) to the considerably higher values achievable with focused pulsed lasers (up to $\sim 10^8$ W/cm^2). Although there is some disagreement over various aspects of the individual processes involved, no doubt due at least in part to the different experimental parameters employed, a great deal has been learned. The work that is described in this paper is intended to further clarify the situation.

Our approach has been to study the fundamental atomic and molecular collision processes that can occur in laser-irradiated sodium vapor by performing experiments at relatively low density $\sim 10^{11}$–10^{13} cm^{-3} using the inherently low power of cw lasers. In this way multiphoton effects could be virtually eliminated, and interactions involving state-selected excited species studied with minimum interference. Using cw lasers we have studied ionization processes[16] such as

$$Na(3p) + Na(3p) \rightarrow Na_2^+ + e \tag{1}$$
and
$$Na(4d) + Na(3s) \rightarrow Na_2^+ + e . \tag{2}$$
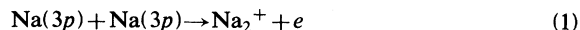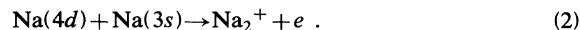
In addition to the associative-ionization channel represented by Eq. (1) we have also studied excitation-transfer processes in energy-pooling $3p$-$3p$ interactions[17] such as

$$Na(3p) + Na(3p) \rightarrow Na(4d) + Na(3s) . \tag{3}$$
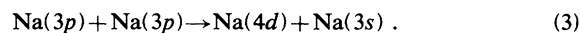
In this earlier work only minor effects were observed if the lasers were not tuned to real atomic resonances. This is doubtless because the low-power density of the cw laser precluded multiphoton effects involving either the atomic or dimer components of the vapor.
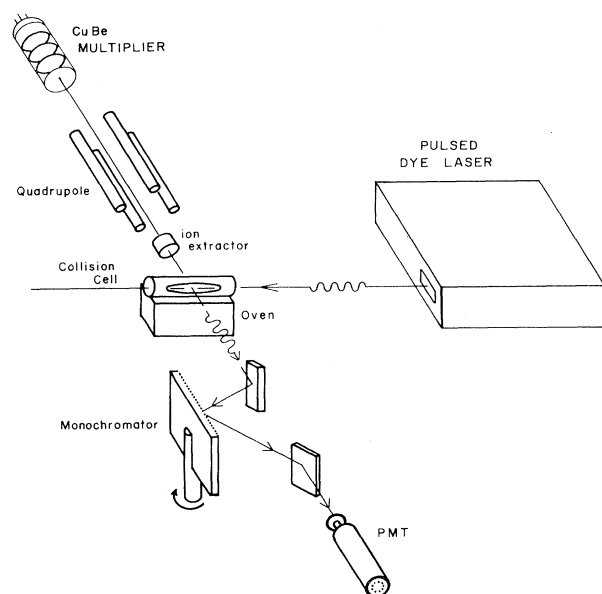
This paper presents the results of experiments on ion

FIG. 1. Schematic diagram of the apparatus.

| | | Bandwidth | |
	Power (W)	GHz	Å
Pulsed dye laser	$\simeq 3.5 \times 10^3$	86	1
cw dye laser			
(multimode)	$\simeq 1.0$	43	0.5
cw dye laser			
(single frequency)	$\simeq 0.12$	0.015	1.5×10^{-4}

centration at these temperatures would be on the order of 1%. However, equilibrium has clearly not been established in this setup so the dimer concentration is likely to be greater than this value.

The pulsed dye laser NRG-Model-DL-03, operated with Rhodamine-6G dye, was pumped with an NRG-Model-0.5 N_2 laser. The laser beam was focused inside the collision cell with a diameter ~ 1 mm. The characteristics of this laser, together with those of the cw lasers used in the related work, are listed in Table I.

Photons emanating from the collision cell were dispersed with a 0.25-m scanning monochromator and detected with a cooled photomultiplier tube (PMT). Ions were extracted from the cell with a set of electrostatic lenses, analyzed by a quadrupole mass filter, and detected with a CuBe particle multiplier. The relative transmission of this system was determined by replacing the oven-cell combination with an electron-impact ion source and comparing the fragment ion yields from various gases with the fragmentation patterns listed in standard tables.[22]

The charge output of either the PMT or the CuBe particle multiplier that resulted from each laser pulse was integrated with a charge-sensitive amplifier and stored in a pulse-height analyzer. After a preset number of pulses (typically 200) the computer read the total charge. Two modes of operation were employed. In one the laser wavelength was fixed and mass scans and/or spectral scans taken. The mass and emission spectra could be acquired simultaneously. In the other mode of operation the settings of the quadrupole mass filter and the monochromater were fixed and the laser wavelength scanned. In either mode the computer controlled operation of the apparatus and stored the data. When photon and ion signals were acquired simultaneously the output of the PMT was fed into a fast counter, the output of which was stored by the computer. Because of the pulsed nature of the experiment this technique does not give reliable absolute photon signals; however, it was used primarily as a wavelength marker.

formation in laser-irradiated sodium vapor using a rather common pulsed laser to investigate higher-power regimes than studied in our previous work using cw lasers. The results demonstrate that dramatic differences occur under otherwise identical experimental conditions when the pulsed laser is substituted for the cw device. In particular, it is shown that the broadband nature of the pulsed-laser light together with the higher-power density effectively magnifies the importance of the dimer concentration so that the ionization process involving dimers obscures ionization from atomic interactions, even when the laser is tuned to a real atomic resonance such as the D line. This is in marked contrast to the cw case where the dimers were found to be virtually unimportant. The data to be presented here strongly suggest that it is two-photon excitation followed by single-photon ionization of the dimers that leads to copious yields of both Na^+ and Na_2^+ at all laser wavelengths employed in this study.

II. EXPERIMENTAL

The experiments reported in this paper were performed with apparatus that is essentially the same as that used previously[17] with a pulsed laser substituted for the cw laser; a schematic diagram is shown in Fig. 1. An oven consisting of a resistance heater imbedded in a Cu block vaporizes the sodium which effuses into the cylindrical collision cell through a slot in one side of the cylinder; 2-mm-diam apertures in the end caps permit entrance and exit of the laser beam. There is a 2-mm$\times$2-cm window on one side for observation of photons and a 5-mm aperture on the other side for extraction of ions. The vapor density in the cell can be varied over the approximate range 10^{11}–10^{13} atoms/cm^3, however, it is difficult to determine the fractional dimer concentration. If thermodynamic equilibrium were established then the dimer con-

III. RESULTS

Mass scans were taken at a variety of different wavelength settings λ_L of the pulsed laser. Figure 2 shows three such scans, each taken with λ_L fixed at a different wavelength, one of which was the D_2 line. It is important to note that the ordinate scale, although arbitrary, is the same for each scan; these data therefore show no enhancement of either the Na^+ or the Na_2^+ signals when the laser is tuned to a D line.

Figure 3 shows data analogous to those shown in Fig. 2, but taken with the cw laser. In contrast to the data ac-
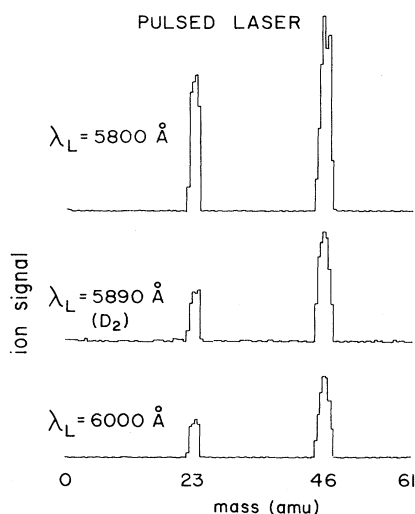
FIG. 2. Mass spectra taken at three different wavelength settings of the pulsed laser. Spectra have been corrected for relative transmission as a function of mass. Ordinates are in arbitrary units, but the three spectra are plotted on the same scale.

quired with the pulsed laser, ions are observed *only* when the cw laser is tuned to a D line. The Na_2^+ ions are from $3p + 3p$ associative ionization,[6,8,9,16] while the small Na^+ signal is from photodissociation[6,15] of the incipient Na_2^+.

The surprising conclusion that the ion yields do not increase when the pulsed laser is tuned to a D line is clearly shown in Figs. 4 and 5. Figure 4 shows the Na^+ and D-line fluorescence signals (taken simultaneously) as functions of λ_L. The axes shown in the figure are included to emphasize the relatively large "background" ion signal at all wavelengths. The D-line fluorescence trace establishes that indeed Na $3p$ are being formed at the appropriate settings of λ_L, and serves as a wavelength marker in the scan. Figure 5 shows traces of the Na_2^+ ion signal and

FIG. 4. Na^+ signal and D-line fluorescence (from both D lines) as functions of λ_L, the wavelength of the pulsed laser. Two sets of axes are shown to indicate the relative magnitudes of the background levels. Both ordinates are in arbitrary units.

$4p \rightarrow 3s$ fluorescence signal (3302 and 3303 Å) as functions of λ_L. No significant enhancement of this ion signal occurs at the D lines, but enhancement of the fluorescence is observed, a result of $3p + 3p$ energy-pooling excitation transfer.[6,9,17] The axes in this figure again indicate that ions are produced at all wavelengths.

Since substantial enhancements of the Na^+ and Na_2^+
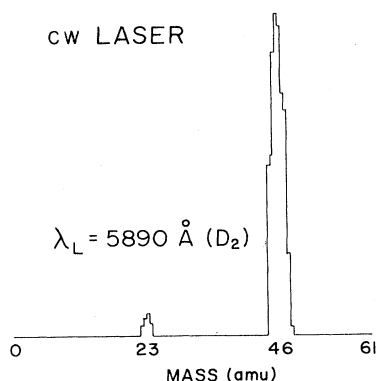

FIG. 3. Mass spectrum taken with a multimode cw laser tuned to the D_2 line. No ions were observed when the cw laser was detuned from a D line. This spectrum, which is shown primarily for comparison with those produced by pulsed laser excitation (Fig. 2), has also been corrected for relative transmission as a function of mass. Ordinate scale is in arbitrary units.
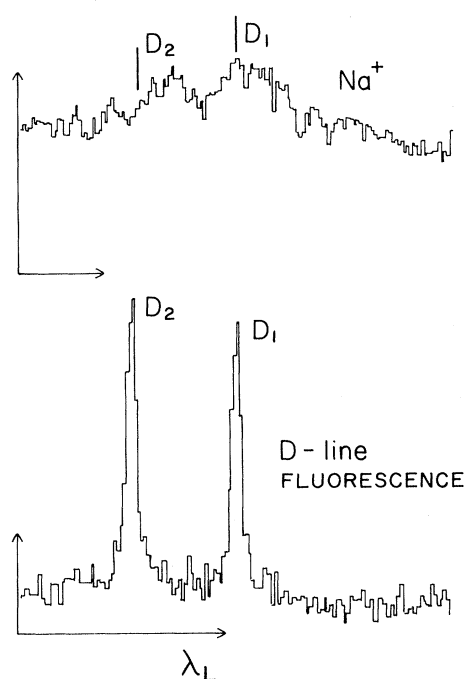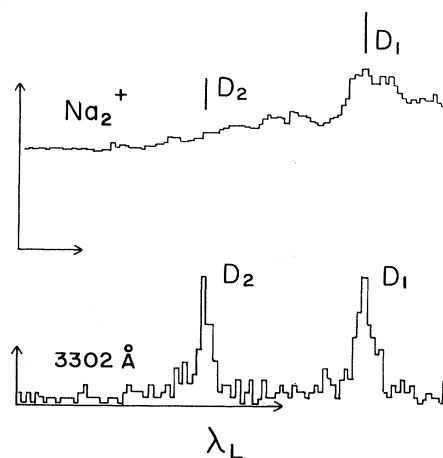

FIG. 5. Na_2^+ signal and $4p \rightarrow 3s$ fluorescence (3302 and 3303 Å) as functions of λ_L, the wavelength of the pulsed laser. Two sets of axes are shown to indicate the relative magnitudes of the background levels. Both ordinates are in arbitrary units.
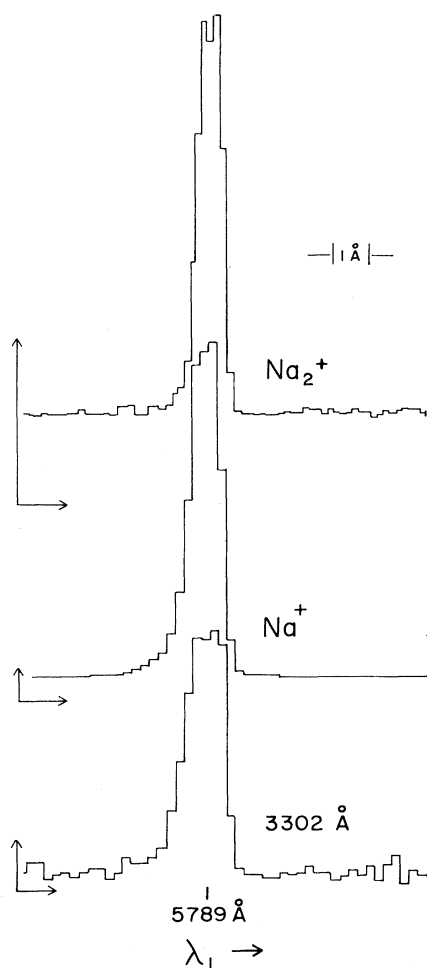
FIG. 6. Na_2^+, Na^+, and $4p \rightarrow 3s$ fluorescence (3302 and 3303 Å) as functions of λ_L, the wavelength of the pulsed laser. Ordinate scales are in arbitrary units, and are unrelated to each other. Three sets of axes are shown to indicate the relative magnitudes of the background levels.
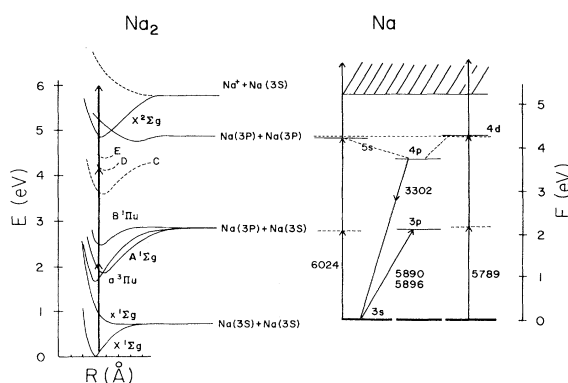

FIG. 7. Partial term diagram for the Na atom showing transitions that can be excited by absorption at the laser wavelengths of these experiments. Also shown in the 3302-Å $4p \rightarrow 3s$ emission detected in the experiments. Dashed line between the $5s$ and $5d$ levels is drawn at twice the energy of the $3p$ state. Also shown are several potential-energy curves of the Na_2 molecule. Vertical arrows on these curves are drawn to indicate the approximate energy of each photon from the excitation laser that may be absorbed by the molecule (each photon has energy in the range 2.05–2.11 eV). Right and left ordinate scales are adjusted so that zero on the right corresponds to Na $3s$ and zero on the left to the bottom of the Na_2 X $^1\Sigma_g$ potential-energy well.

signals were expected when the pulsed laser was tuned through the D lines it was considered important to find some condition under which ion peaks could be observed as λ_L was varied in order to establish proper operation of the apparatus. Assurance of proper operation was achieved by tuning the pulsed laser through either the $3s \rightarrow 4d$ or $3s \rightarrow 5s$ two-photon resonances at 5789 and 6024 Å, respectively. Figure 6 shows the Na^+, Na_2^+, and 3302-Å signals as λ_L is scanned through a very small range that includes the $3s \rightarrow 4d$, two-photon atomic resonance. The Na^+ ions are from three-photon resonance-enhanced ionization; the origin of the Na_2^+ will be discussed in Sec. IV. The 3302-Å $4p \rightarrow 3s$ fluorescence is a result of the cascade from the $4d$ state via the $4d \rightarrow 4p$ infrared transition. These data unequivocally establish that the apparatus is indeed operating properly. Similar observations were made as the pulsed laser was scanned

through the 6024-Å, $3s$-$5s$, two-photon atomic resonance.

IV. DISCUSSION

The laser wavelengths used in these experiments, 5750–6050 Å, make it possible to access a number of Na and Na_2 states by both one- and two-photon absorption. This is illustrated in Fig. 7 which shows a partial term diagram for Na and potential-energy curves for several Na_2 states. Some of the transitions that can be excited with laser light in the range of wavelengths used in this work are shown in the figure. Also shown are the transitions from the $4d$ level that lead to 3302-Å radiation by cascading.

A number of two-photon absorptions in Na_2 have recently been reported[23–27] for the same wavelength range as that employed in our experiments. These two-photon absorptions can serve as the first step in resonance-enhanced three-photon ionization. We believe that these resonances are important steps in the multiphoton ionization of Na_2 to both bound and repulsive states of Na_2^+. This conclusion is consistent with one reached by Wu et al. in a recent paper.[28]

The most unexpected feature of the data presented here is the absence of significant enhancement of either ion signal when the pulsed laser is tuned to a D line. Enhancement of the Na^+ signal might have resulted from three-photon resonance-enhanced ionization of sodium atoms, and that of Na_2^+ from $3p + 3p$ associative ionization. Obviously these two processes are occurring, however, because the Na_2 molecules absorb at essentially all wavelengths in this range, including those corresponding to atomic resonances; Na^+ and Na_2^+ are copiously produced

from the dimer. These ions are presumably obscuring the Na^+ and Na_2^+ that result from interactions involving atoms when the laser is tuned to a D line. The fact that dimer molecules lead to ion formation with the pulsed laser, but not with the relatively low-power cw laser, suggests that two-photon excitation of high-lying molecular states, via virtual intermediate states, rather than sequential excitation, via a real intermediate state, is the first step in the multiphoton ionization of the dimer. Although significant enhancement of the ion signals was not observed at the D line we do see relatively large increases in both Na^+ and Na_2^+ at the $3s \rightarrow 4d$ and $3s \rightarrow 5s$ two-photon atomic resonances. The increased Na^+ signal is almost certainly from three-photon resonance-enhanced ionization via the $4d$ (or $5s$) level, a process that is expected to be orders of magnitude larger than three-photon resonance-enhanced ionization via the $3p$ level.[29] In fact, our data confirm this.

As noted above it was anticipated that $3p + 3p$ associative ionization would cause the Na_2^+ signal to increase dramatically as the laser was scanned through the D lines. Unlike three-photon ionization, associative ionization requires a heavy-body—heavy-body collision. Since the heavy bodies in this case must both be $Na\,3p$, associative ionization can occur only during the time interval in which these excited atoms are present. Because the laser pulse is shorter than the 16-nsec natural lifetime of the $3p$ state it is the lifetime of this state that dictates the temporal dependence of the potential reactants. (If radiation trapping is considered then the expanding reaction volume must also be taken into account so that this complication will be neglected for the purpose of this discussion.) Because significant enhancement of the Na_2^+ signal is not observed at the D lines it is apparent that the cross section for $3p + 3p$ associative ionization[8,16] ($\sim 10^{-17}$ cm^2) is insufficient to produce an Na_2^+ signal (during the time that $Na\,3p$ are present) that is measurable above the ion background from processes involving the dimers. This is in contrast to our earlier work with cw lasers, in which the dimers were found to be unimportant. Recall that no signals, photon or ion, were observed when the cw laser was tuned off a D line so there was no background signal from dimer ionization. Furthermore, in the cw case there is a continuous supply of potential reactants, the $Na\,3p$ atoms.

We do, however, observe enhancement of the Na_2^+ signal as the pulsed laser is scanned through the two-photon atomic resonances. There are two possible sources of these increased Na_2^+ yields. Since the ionization potential of atomic sodium (5.1 eV) is greater than the ionization potential of the dimer (4.9 eV), charge exchange in Na^+-Na_2 collisions can contribute to the Na_2^+ yield. If this is the case then the increased Na^+ concentration at these wavelengths is directly responsible for the observed Na_2^+ increase. To our knowledge there are no data available on this charge-transfer process.

A second possible source of Na_2^+ at the two-photon atomic resonances is $3s + 4d$ or $3s + 5s$ associative ionization.[16] Although $3p + 3p$ associative ionization could not be detected above the dimer-produced Na_2^+ signal, there are several factors that could permit the observation of $3s + 4d$ and $3s + 5s$ processes, especially the former. First, the lifetimes of the $4d$ and $5s$ states are longer than that of the $3p$ state, 55 and 80 nsec, respectively.[30] Second, in

each of these procedures, $Na\,3s$, which is the major component of the vapor, is one of the reactants and is of course continuously present. Thus, although the $4d$ or $5s$ concentration is no doubt lower at the respective laser wavelengths than is the $3p$ concentration at the D lines, the significantly larger concentration of $Na\,3s$, together with the longer lifetimes, may conspire to make these associative-ionization processes observable while that from $3p + 3p$ collisions is not.

We have also observed that the ion signal due to the dimers tends to decrease with increasing wavelength. That is, the dimer-produced background is greater at the $3s \rightarrow 4d$ wavelength 5789 Å than at the D lines (or the $3s \rightarrow 5s$ wavelength 6024 Å). Although this background is higher, by about a factor of 2, the $3s + 4d$ associative-ionization cross section is about 60 times larger[16] than that for $3p + 3p$. However the $3s + 5s$ cross section is about 40 times lower[16] than that for $3p + 3p$; yet an enhancement of the Na_2^+ signal is observed at 6024 Å. These observations suggest that Na^+-Na_2 charge transfer discussed above plays a major role in the production of Na_2^+ at the wavelengths of the two-photon atomic resonances, especially the $3s \rightarrow 5s$ transition.

The results presented here are in contrast to those of Roussel et al.[12] who have reported data for Na_2^+ production as a function of pulsed-laser wavelength at vapor densities comparable to those employed in our experiments. While, in agreement with our data, they observe an enhancement of the Na_2^+ signal at 6024 Å (they did not report data for λ_L as short as 5789 Å), they also observe significant enhancement of the Na_2^+ signal at the D lines. The major difference between their experiment and ours is that they used a flashlamp pumped pulsed laser, for which the pulse length is ~ 1 μsec, approximately 100 times longer than that of the laser used in our experiments. Thus it seems likely that this pulse length permits the active medium for production of Na_2^+ by associative ionization, the $Na\,3p$ atoms, to survive for a sufficient time to permit reaction.

The observation by Lucatorto and McIlrath[1] that a column of Na vapor could be completely ionized when irradiated by ~ 1 MW of laser light tuned to a D line has been attributed to ionization by superelastically heated electrons,[31-35] electrons that have acquired kinetic energy in e-$Na\,3p$ quenching collisions. These hot electrons are then capable of ionizing $Na\,3p$ or even $Na\,3s$. Each ionization event adds to the electron inventory so that a cascading process results. It has been proposed that the "seed" electrons for this avalanche are provided by $3p + 3p$ associative ionization,[8,9,18,35] so that our data, together with those of Roussel et al.,[12] suggest that the use of a flashlamp pumped laser may have been an important aspect of the experiments of Lucatorto and McIlrath.[1] However, based on our data, it seems likely that even if a laser of much shorter pulse duration, such as one that is pumped by either an N_2 or a Q-switched Nd:YAG laser, is used, seed electrons could be produced by ionization of the dimer component of the vapor. Such effects would be magnified at the higher densities employed by Lucatorto and McIlrath because the fractional dimer concentration is higher than in our experiments. In any case our data show that dimer ionization contributes substantially to the electron concentration in laser-irradiated vapor.

Although we were unable to detect Na_2^+ from $3p + 3p$ collisions above the dimer-produced background, our observation of 3302-Å radiation as the pulsed laser was scanned through the D lines shows clearly that $3p + 3p$ energy-pooling excitation transfer is occurring. Previously we have shown that the primary products of such excitation transfer are Na $4d$ and Na $5s$,[17] and that the 3302-Å radiation is indicative of the formation of these states. Given that we were unable to detect $3p + 3p$ associative ionization in this work how then were we able to detect $3p + 3p$ excitation transfer? Of course, the relative magnitudes of the cross sections for each of these processes is important, but there is disagreement in the literature over the cross sections for $3p + 3p$ excitation transfer.[14,16,20,21] The reported values range from about an order of magnitude larger than our previously measured $3p + 3p$ associative-ionization cross section to about three orders of magnitude lower than the associative-ionization cross section. However, from an experimental point of view the reason that 3302 Å can be measured, while Na_2^+ produced by associative ionization cannot, is simply that there is little or no 3302 Å background, but the dimer-produced ion signal is sufficient to obscure the Na_2^+ signal from $3p + 3p$ associative ionization. The data in Fig. 5 illustrate this point.

Finally, it is worthwhile to consider the mechanisms by which the dimer component of the vapor produces so many ions. As discussed previously it is difficult to reliably estimate the dimer fraction of the vapor due to the nonequilibrium nature of the reaction cell. Our data suggest however that this fraction may be considerably higher than 1%, the fraction calculated under the assumption of thermodynamic equilibrium. Nevertheless, while only a small fraction of the photons from a broadband laser, such as the one used in this work, can excite an atomic transition, virtually all of the photons in the laser beam can excite Na_2. This effectively magnifies the Na_2 concentration

relative to that of the atoms so that the ratio of excited Na_2 molecules to excited atoms can be significantly greater than the ratio of ground state species.

Wu and Judge, in collaboration with Roussel and co-workers,[28] have reached the conclusion that in the earlier work of Roussel et al.[12] the production of Na_2^+ at laser wavelengths that do not coincide with atomic resonances was due primarily to the dimers. This conclusion is of course consistent with the main theme of the discussion in this paper. They suggest that Na_2^+ results from two-photon absorption by the Na_2 molecules, followed by single-photon photoionization, as we suggest in this paper. Further, Muller and Hertel[4] suggest that two-photon excitation of the dimers is the primary excitation mechanism that leads to the infrared laser emission in their experiments.

It is possible that ions are being produced by interactions that are driven by the presence of the strong electromagnetic field associated with the laser, so-called laser-induced reactions. This possibility was precluded in the cw experiments, but it must be considered when pulsed lasers are employed. Although there have been some recent experiments on laser-induced processes in sodium vapor,[11−13,36] the picture is unclear at this time. Despite the potential complications introduced by the presence of neutral dimers, studies of laser-induced effects in sodium vapor are continuing[37,38] in several laboratories, including our own.

ACKNOWLEDGMENTS

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