Polarization in Collisions between Ultra-cold Sodium Atoms

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Abstract. We have studied polarization effects in the photoassociation of two colliding ultra-cold Na-atoms. In a first step the two colliding atoms absorb a laser photon to associate to a singly excited molecule in a bound state of the 0_g^- or 1_g potential. In a second step the excited molecule absorbs a second photon and is further excited to a bound level in a doubly excited potential (0_u^- or 1_u which are autoionizing). By measuring the produced ions with parallel and perpendicular polarizations of the two laser beams used possible polarization effects can be studied. We find strong polarization effects if a 0_g^- state is used as the intermediate state and very little or no effects at all if a 1_g state is used.

INTRODUCTION

The study of collisions between cold atoms (T < 1 mK, corresponding to relative velocities of the order of a few m/s) has been made possible by the introduction of laser cooling and trapping techniques. When two very slow atoms collide in the presence of a radiation field they may combine to produce a bound, electronically excited molecule. This photoassociation process may be detected as a decrease of the fluorescence of the trapped atomic sample [1]. Alternatively the excited molecule may be excited by a second photon to a higher lying state which lies in the ionization continuum or which is autoionizing via a crossing with the continuum at short internuclear range. By measuring the ions formed as a function of the frequencies of the lasers both the first and the second step of this process may be studied. This is shown schematically in the Figs. 1a and 1b, respectively. In the recent past several experiments of this type have been performed and valuable information has been obtained on the potential curves of the states involved [2, 3].

In the present experiment we have studied the above described two-step process as a function of the polarization of the two lasers used. When a sample of isotropically oriented molecules is excited by polarized light the excited molecules will in general be aligned with respect to the polarization direction of the exciting light beam [4]. This is a consequence of the fact that the absorption probability is dependent on the orientation of the molecular axis with respect to the polarization direction of the light. Therefore, if a sample of cold atoms moving with random relative velocity directions is irradiated with polarized light, atom pairs with favorable relative velocity directions will preferentially absorb a photon and combine to an excited molecule. Immediately after its formation this molecule will have its axis aligned along the relative velocity direction of the two colliding atoms before the absorption. Similarly the probability for the second photon to



FIGURE 1. Schematic diagram showing ground, singly-excited and doubly-excited potentials of the Na_2 molecule. Two ground state atoms colliding slowly absorb a red detuned photon to produce a singly-excited, translationally cold molecule in a well-defined rovibrational level. By absorption of a second photon the singly-excited molecule may be further excited to a doubly-excited (autoionizing) state. By monitoring the ion production as a function of the frequency of the first (left panel) or second (right panel) photon, the rovibrational levels in the singly- or doubly-excited potential may be probed, respectively.

be absorbed in the second step will depend on the relative orientation of the molecular axis in the intermediate excited state with respect to the polarization direction of the second photon. As a result the production rate of the ions formed in the autoionization process is expected to depend on the relative orientation of the polarization directions of the two successive photons that have been absorbed.

In our experiment we have chosen as the intermediate state the (v, J) = (1, 2) rovibrational level in the 0_g^- or 1_g potential which asymptotically connect to the $S_{1/2} + P_{3/2}$ dissociation limit of Na₂. The final state reached upon the second photon absorption is one of the bound levels in either the 0_u^- or the 1_u potential connecting to the $P_{3/2} + P_{3/2}$ dissociation limit.

EXPERIMENT

The experiments are performed in a so-called "dark-spot" magneto-optical trap (MOT) [5] which leaves almost all atoms in the lowest, f = 1, hyperfine state. The trap is continuously loaded from a Zeeman-tuned, laser cooled, atomic beam [6]. The trapping beams are turned on and off at a 100 kHz rate with a duty cycle of 50%. During the trap off periods two probe beams are focused in the MOT at the region with the highest density of trapped atoms. One of the lasers (the PA-laser) is detuned to the red of the atomic $S_{1/2} + P_{3/2}$ resonance while the other laser (the PI-laser) is detuned to the blue. The frequency of the PA-laser is adjusted to drive a transition from the colliding ground state atoms to a specific rovibrational level in the 0_g^- or 1_g potential. The frequency of the PI-laser is scanned to drive the transitions from the chosen inter-



FIGURE 2. Spectra of rovibrational lines within the highest two vibrational bands in the 0_u^- and 1_u doubly-excited potentials. The rovibrational levels are excited via the two-step mechanism depicted in Fig. 1b with the PA-laser fixed at the transition to the (v, J) = (1, 2) rovibrational level in the 0_g^- potential. The laser light is linearly polarized, where in spectrum (a) the polarizations of the PA- and PI-laser are parallel to each other and in spectrum (b) they are perpendicular to each other. The baseline of spectrum (b) has been shifted for clarity. Both spectra have been normalized to each other by setting the intensity for the $0_u^-(J=3)$ peak equal to 100.

mediate state to bound levels in the 0_u^- or 1_u potentials which are autoionizing at short internuclear range. By collecting the produced ions an ion spectrum is measured where the peaks correspond to the bound levels in the doubly excited potentials. The whole process is shown schematically in Fig. 1b. To facilitate the study of polarization effects the light of the PA- and PI-lasers is linearly or circularly polarized. The ion spectra are measured in two cases: (i) With the (linear or circular) polarizations oriented parallel to each other and (ii) With the polarizations oriented at right angles to each other.

RESULTS

Figure 2 shows the results of our measurements when linearly polarized light is used for the two successive excitation steps. The frequency of the first (PA) laser is fixed at the transition to the (v, J) = (1, 2) rovibrational level in the 0_g^- potential, whereas the second (PI) laser is scanned through the bound levels in the 0_u^- or the 1_u doubly excited potentials. The identifications of the various lines in the spectra have been made using the dipole selection rules. In the spectra we have identified two vibrational levels of each molecular symmetry, 0_u^- and 1_u , as indicated. Within each vibrational band



FIGURE 3. Same as Fig. 2, but now with the laser light circularly polarized.

rotational lines are observed with quantum numbers J = 1, 2 or 3. (Remember that the intermediate state is a (v, J) = (1, 2) level). For the vibrational levels of the 0_u^- state the J = 2 rotational line is clearly missing. This is a result of the selection rule $\Delta J \neq 0$ for a $J = 0 \rightarrow J' = 0$ transition. For some hitherto unexplained reason the J = 1 line of the second vibrational band of the 0_u^- state seems to have split in two lines. In panel (a) the two polarizations are oriented parallel and in panel (b) they are oriented at right angles to each other. When comparing the two panels in Fig. 2 it is immediately clear that the relative intensities of the various peaks differ appreciably in the two cases, indicating that strong polarization effects are present. Similar results have been obtained for circular polarization (see Fig. 3). Also in that case strong polarization effects are observed. The results have been summarized in Table 1. All spectra have been normalized to each other by setting the intensity of the 0_u^- (J = 3) peak equal to 100. This strong peak appeared to vary negligible when the orientation of the two polarizations was changed from parallel to perpendicular. In the column 6 and 9 of the table the ratios of the peak intensities for parallel and perpendicular orientation of the two polarizations are given for linear and circular polarization, respectively. We see that for transitions from the intermediate to the final state with $\Delta J = +1$ (R-branch) the polarization effect is small (the parallel/perpendicular ratio ≈ 1) whereas for transitions with $\Delta J = 0$ (Q-branch) and $\Delta J = -1$ (P-branch) they are strong. The strange J = 1 states in the 0_u^- potential appear to deviate in this respect.

Similar experiments as described above have been performed, but with one of the rovibrational levels of the 1_g state chosen as the intermediate state. In this case very little or no polarization effects at all were observed.

TABLE 1. Intensities of the rotational levels within the highest two vibrational bands of the 0_u^- and 1_u symmetry. For each polarization combination the intensities are normalized with respect to the $\Delta J = +1$ transition of the highest vibrational state of the 0_u^- symmetry. The intensity of this peak appeared to change negligible, when the orientation of the polarizations was changed from parallel to perpendicular. The results are averages over three scans for each combination.

v	state	ΔJ	lin lin	lin⊥lin	$\lim \ \ln/\ln \perp \ln$	$\sigma\!-\!\sigma$	$\sigma^+ - \sigma^-$	$\sigma\!-\!\sigma\!/\sigma^{\!+}\!-\!\sigma^{\!-}$
$v_{\rm max} - 1$	0_{μ}^{-}	-1	1.06	4.78	0.22	0.28	2.79	0.10
		+1	6.60	6.45	1.02	2.51	2.21	1.13
$v_{\rm max} - 1$	1_u	-1	0.16	2.04	0.08	0.16	3.98	0.04
		0	1.89	20.53	0.09	0.05	8.95	0.006
		+1	23.02	24.48	0.94	9.18	10.11	0.91
$v_{\rm max}$	0_u^-	-1	24.28	59.66	0.41	9.49	77.86	0.12
		-1	39.25	70.60	0.56	14.06	119.08	0.12
		+1	100	100	1	100	100	1
$v_{\rm max}$	1_u	-1	6.93	26.16	0.26	1.12	25.93	0.04
		0	14.38	55.11	0.26	6.69	54.69	0.12
		+1	23.82	23.56	1.01	13.06	16.48	0.79

DISCUSSION

At first sight it is surprising that polarization or orientation effects do exist at all for the above described processes. Since the rotational spacing is of the order of 1 GHz the excited molecule in the intermediate state may make several rotations during its lifetime which is of the order of 10 ns. In this case one would expect the electronic angular momentum precessing around the molecular axis to have completely lost all memory of its initial orientation by the time of the second photon absorption. However, this is only true if the electronic angular momentum indeed precesses around the molecular axis. The 0_g^- state has zero angular momentum component along the molecular axis and hence there is no rotation around this axis. Therefore the electronic angular momentum remains fixed in space during the rotation of the molecule at right angles with the plane of rotation. The 1_g state on the other hand does have an electronic angular momentum component along the molecular axis and therefore the electronic angular momentum vector carries out precessions around the rotating axis and the memory of its initial orientation is lost by the time of the second photon absorption. This is most likely the reason why we fail to observe significant polarization effects when we choose one of the rovibrational states in the 1_g potential as the intermediate state.

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