Polarization dependence of $Na^* + Na^*$ associative ionization revisited

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The dependence of the associative ionization process $Na\ 3^2P_{3/2} + Na\ 3^2P_{3/2} \rightarrow Na_2^+ + e^-$ on the polarization of the laser light used for Na excitation was independently investigated in Utrecht and Berlin. The purpose of this paper is to clarify discrepancies between earlier experimental results of Kircz, Morgenstern, and Nienhuis, on one hand, and Rothe, Theyunni, Reck, and Tung, on the other hand. The new results confirm in general the data of Kircz, Morgenstern, and Nienhuis, and also indicate a dependence of the anisotropy ratios on the relative velocity of the interacting Na^* atoms.

In the past the polarization dependence of associative ionization in $Na^* + Na^*$ collisions has been investigated independently by Kircz, Morgenstern, and Nienhuis[1] and by Rothe, Theyunni, Reck, and Tung.[2] In both cases Na atoms of a single effusive beam were excited with linearly polarized light of a dye laser, tuned to the $F = 2 \rightarrow F' = 3$ hyperfine transition of the Na D_2 line. Due to their Maxwellian velocity distribution the atoms could collide, the direction of their relative velocity being preferentially in the direction of the Na beam. When the angle θ between the polarization vector of the laser light and the atomic beam was varied, both groups observed a significant variation of the total ionization signal that is due to associative ionization. It can be shown[1,3] that the ionization signal depends on the angle θ according to

$$I = 1 + r_1 \cos(2\theta) + r_2 \cos(4\theta) \quad . \tag{1}$$

Whereas both groups[1,2] could fit their measured data to this functional form, discrepancies occurred regarding the coefficients r_1 and r_2. Kircz *et al.*[1] found $r_1 = 0.27$ and $r_2 = 0.10$, whereas Rothe *et al.*[2] found $r_1 = 0.01$ and $r_2 = 0.38$. These discrepancies initiated a more detailed investigation of the $Na^* + Na^*$ associative ionization in Utrecht and in Berlin in order to find out how far different experimental conditions can influence the observed results.

In Utrecht the original experimental setup of Kircz *et al.*[1] was modified in two respects. First of all, the ion detection system was redesigned such that resonance fluorescence light from Na^* could no longer reach the particle multiplier. In this way a possible influence of fluorescence light on the Na_2^+ detection efficiency could be excluded. Rothe *et al.*[2] had discussed such an influence as a possible source of error. In the original setup the multiplier was placed at 90° with respect to the directions of atomic and laser beam. In this direction the emitted fluorescence light shows a similar dependence on θ as the ion signal we observed. In addition, we did, of course, all tests which had also been done in the original experiment: We checked the polarization properties of the laser light at various places, we monitored the fluorescence intensity as a function of θ, we varied size and shape of the interaction region, and we checked that the ionization signal disappeared when a retarding field was applied in the interaction region. However, the experimental results with our new setup did not differ significantly from the former ones.

As another modification we installed a second atomic Na beam propagating exactly in opposite direction with respect to the first beam. By doing so the number of collisions was increased significantly. Furthermore, we can now distinguish between two types of collisions: (i) "head-tail" (h-t) collisions of atoms within one of the beams at low kinetic energies and (ii) "head-head" (h-h) collisions between atoms from different beams at higher kinetic energies. The corresponding ionization signals can experimentally be identified by shutting off the various beams in a suitable sequence. The results are shown in Fig. 1. The ionization signal is normalized to 1 at $\theta = 0°$. At oven temperatures of $T_1 = 635$ K and $T_2 = 550$ K we calculate[4] mean relative collision velocities of $\bar{v} = 520$ m/s and 1565 m/s for h-t and h-h collisions, respectively. In both cases we clearly observe a 180° periodicity of the ionization signal. The polarization dependence is even more pronounced at higher collision energies. The velocity dependence of the ionization cross section will be reported in a forthcoming publication.[5] A fit of the curves with the functional dependence (1) yields r values that are given in Table I.

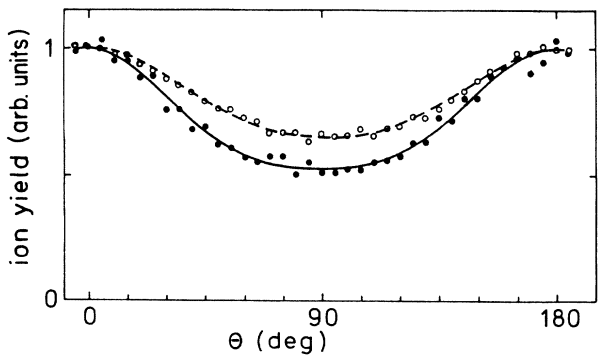

FIG. 1. Polarization dependence of the ion yield of the process $Na\ ^2P_{3/2} + Na\ ^2P_{3/2} - Na_2^+ + e^-$, Utrecht measurements with best fit, for ●—● $\bar{v} = 1565$ m/s (h-h collisions), ○--○ $\bar{v} = 520$ m/s (h-t collisions).

TABLE I. Values of the fit coefficients r_1 and r_2 of (1) (for the 2θ and 4θ dependence, respectively), and the corresponding mean collision velocity $\bar{v}$.

	r_1	r_2	$\bar{v}$ (m/s)	
h-t	0.225 ± 0.004	0.032 ± 0.003	520	Utrecht
h-h	0.304 ± 0.006	0.0664 ± 0.0024	1565	
h-t	0.239 ± 0.005	0.010 ± 0.004	750	Berlin

The experiment in Berlin was performed in a single atomic beam generated by a two chamber recycling oven and employing a standard experimental setup used in this type of polarization studies with laser-excited atoms.[6] The Na_2^+ ions were extracted from the optical excitation region by an electric field and detected by a system consisting of field electrodes and a particle multiplier. By applying a retarding field in the detector we found that the Na^* fluorescence did not simulate an ion signal on the multiplier. Linear polarization of the laser light was verified before entering the apparatus as well as in the laser beam transmitted through the entire experimental setup. In order to monitor the optical pumping process and the alignment of the excited atoms the fluorescence from the excitation region was measured simultaneously with the ion signal, a precaution which appears crucial in such polarization studies.

The measured ion yield from the associative ionization is shown in Fig. 2, together with the fluorescence as a function of the laser polarization angle θ with respect to the Na beam direction. The fluorescence shows the maximum anisotropy to be expected, for maximum alignment of the $Na(3^2P_{3/2})$ atoms.[7] In our case the fluorescence detector is positioned at 45° with respect to the plane in which the polarization vector is rotated.

In order to determine the average relative mean collision velocity in the Berlin Na beam, we have measured Doppler spectra of the laser-induced fluorescence which yield the velocity distribution. Figure 3 displays the experimental results [corrected for a background from the second hyperfine component of the $Na(3^2S_{1/2})$ ground state] and for comparison the Maxwellian velocity distribution (broken curve) based on the nozzle temperature of $T = 910$ K. Though the oven temperature is only $T = 700$ K and the nozzle diameter is $D = 0.5$ mm, our atomic beam is already in the transition region from effusive to hydrodynamic behavior.

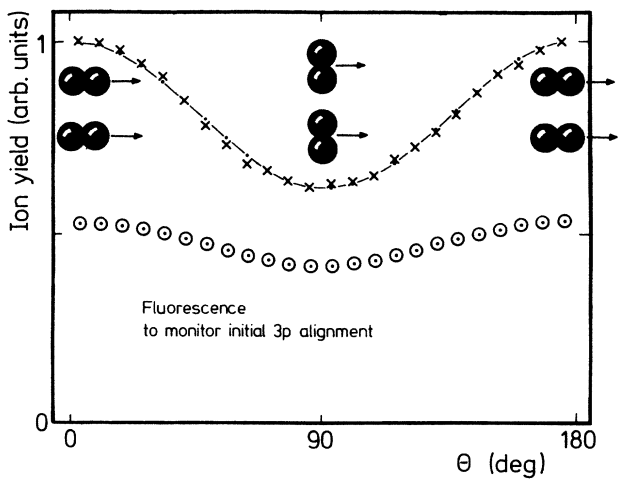

FIG. 2. Polarization dependence of associative ionization (same as Fig. 1), Berlin measurements with best fit, $\bar{v} = 750$ m/s.

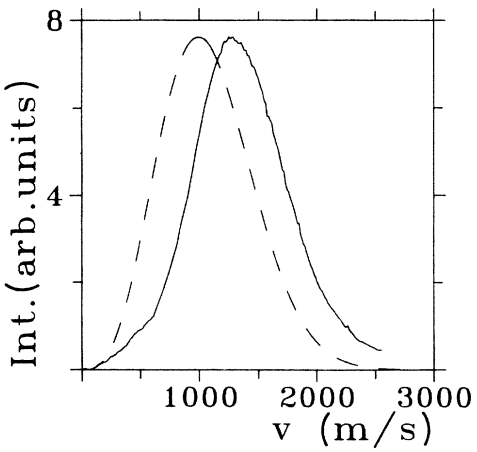

FIG. 3. Velocity distribution of the Berlin sodium atomic beam measured by Doppler-shifted laser-induced fluorescence. The broken curve displays the calculated Maxwellian velocity distribution for our nozzle temperature $T_{nozzle} = 910$ K.

The r values of the Berlin polarization measurements are also shown in Table I. Obviously, the r_1 values increase with the collision energy. In contrast the (small) r_2 values appear to have a more complicated behavior. This may be rationalized by a comparison with measurements of the total cross section of associative ionization[8] (without polarization analysis) which shows also a complicated energy dependence.

Since the new experiments in Utrecht and Berlin both confirm the original results of Kircz *et al.*,[1] we would like to

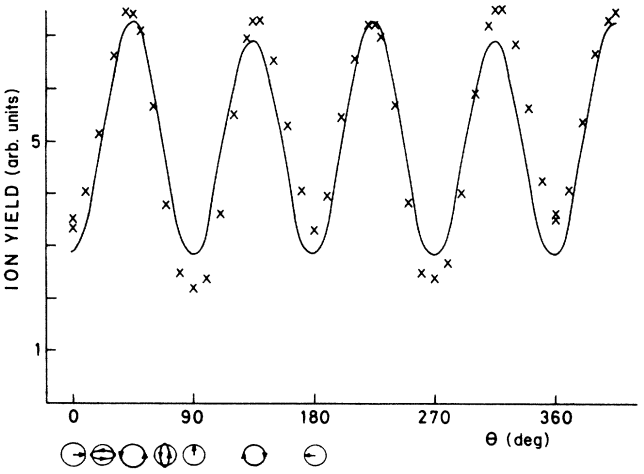

FIG. 4. Polarization dependence of the ion signal with a $\lambda/4$ plate in the laser beam, as measured in Utrecht. The polarization is shown below the figure. The main axes of the $\lambda/4$ plate are at 0° and 90°. Therefore, at those angles linear polarized light is transmitted, whereas at other angles the light is elliptically polarized. At 45° circularly polarized light is transmitted. × represents experimental points, —— represents best fit with (1), with $r_1 = 0.04 \pm 0.04$, $r_2 = 0.43 \pm 0.03$. The laser irradiance was 35 mW/cm^2, the size of the spot was 5×7 mm^2.

close with a discussion of possible reasons for the different results that Rothe *et al.*[2] had found initially. One possible explanation might be that in those experiments a considerable amount of circularly polarized light is produced at some angles θ. Such an admixture would severely influence the data. It could be induced by birefringent properties of the windows that the laser light has to pass on its way to the vacuum chamber. Such birefringence could, e.g., be caused by mechanical tensions. By means of circularly polarized light a much higher density n of excited atoms can be obtained than by linearly polarized light since in a stationary situation only $M_F = 2 \rightarrow M_{F'} = 3$ transitions are induced, thus excluding excitation of the $F' = 2$ state that can decay to the "wrong" hyperfine level $F = 1$ of the ground state.[7] At the laser powers and beam diameters used by Rothe *et al.*[2] this is a considerable effect. Since the ionization signal due to collisions within one beam is proportional to n^2, it could be increased significantly in this way. With a "birefringent" window one would expect a corresponding increase of ionization with a periodicity of 90° in θ, since every 90° the polarization vector would be halfway between the main axes of the birefringent window. To imitate such an effect in an exaggerated way we have placed a $\lambda/4$ plate in front of our window and have repeated the experiment. The result is shown in Fig. 4.

Indeed, now we observe maxima of ionization every 90° such as Rothe *et al.*[2] The intensities of these maxima are significantly higher than for the cases of linearly polarized light. Another important point is the fact that every second minimum is lower or higher than its neighboring minimum. As indicated at the bottom of Fig. 4, the minima occur for the case of excitation with linearly polarized light, however, with a true periodicity of 180°, since the polarization vectors for neighboring minima are perpendicular with respect to each other. Therefore, they represent a different situation regarding the atomic collision velocity **v**. A closer inspection of the data of Rothe *et al.*[2] shows that indeed the minima are alternately low or high. This supports our suspicion that admixtures of circularly polarized light might have influenced their data. Recent results of Rothe, Theyunni, Reck, and Tung[9] are in close agreement with the data presented here and support the explanation given above.

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[1] J. G. Kircz, R. Morgenstern, and G. Nienhuis, Phys. Rev. Lett. **48**, 610 (1982).

[2] E. W. Rothe, R. Theyunni, C. P. Reck, and C. C. Tung, Phys. Rev. A **31**, 1362 (1985).

[3] G. Nienhuis, Phys. Rev. A **26**, 3137 (1982).

[4] The mean relative velocity given here is calculated from the density distributions $n(v)$ as

$$\bar{v} = \frac{\int\int (v_1 - v_2)^2 n_1(v_1) n_2(v_2) dv_1 dv_2}{\int\int |v_1 - v_2| n_1(v_1) n_2(v_2) dv_1 dv_2} ,$$

the integrand of the denominator giving the probability for measuring a relative velocity $|v_1 - v_2|$ in a collision.

[5] H. A. J. Meijer, H. P. v. d. Meulen, R. Morgenstern, and G. Nienhuis (unpublished).

[6] A. Bähring, E. Meyer, I. V. Hertel, and H. Schmidt, Z. Phys. A **320**, 141 (1985).

[7] A. Fischer and I. V. Hertel, Z. Phys. A **304**, 103 (1982).

[8] M.-X. Wang, M. S. de Vries, J. Keller, and J. Weiner, Phys. Rev. A **32**, 681 (1985).

[9] E. W. Rothe, R. Theyunni, C. P. Reck, and C. C. Tung, this issue, Phys. Rev. A **33**, 1426(E) (1986).