# Influence of Coherence on Associative Ionization in Na(3$p$) + Na(3$p$) Collisions

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(Received 14 September 1987)

We have investigated the associative ionization of Na atoms in two counterpropagating thermal beams independently excited by various combinations of linearly and circularly polarized light. In this way information about coherence contributions to the ion signal has been obtained for the first time. These contributions which correspond to off-diagonal elements of the detection matrix have hitherto been neglected, but are of the same magnitude as the cross sections themselves and depend on the velocity.

PACS numbers: 34.50.Lf, 34.50.Fa, 82.40.Dm

In recent years many groups have investigated associative ionization occurring in collisions between two laser-excited Na(3$p$) atoms,

$$\mathrm{Na}(3p)\,^2P_{3/2}+\mathrm{Na}(3p)\,^2P_{3/2}\rightarrow \mathrm{Na_2}^+ + e^-, \qquad (1)$$

especially after Kircz, Morgenstern, and Nienhuis[1] had found a strong dependence of this reaction on the polarization of the collision partners. Since that time several groups[2-7] have measured this polarization dependence by exciting the Na atoms with linearly or circularly polarized light in various configurations. One problem in these experiments is the difficulty in preparing the collision partners in simple atomic states. The situation before the collision has to be described by density matrices, accounting for the atomic states and the different directions of the laser polarization with respect to the direction of the atomic beams. For the comparison of different experimental results with each other and with theory, it is extremely important to have well-defined excitation conditions in order to allow an unraveling of the measured signals, e.g., in terms of ionization cross sections for atoms colliding with each other in well-defined substates. Such an unraveling was not possible for all experiments and so far has only been performed in a few cases.[1,5-7]

In the analyses it was hitherto assumed that coherence between the various excited Na states has no influence on the ionization. This assumption, up to now enforced by the lack of sufficient experimental information, implies that the measured signals can be completely described by magnetic-sublevel-dependent ionization cross sections $\sigma(m,n)$, characterizing collisions with one atom in sublevel $m$ and the other in sublevel $n$. The $\sigma(m,n)$ can be written as squares of ionization amplitudes $|f(m,n)|^2$, and the question is to what extent it is justified to neglect the coherence terms $f^*(m,n)f(m',n')$, with $(m,n)\neq(m',n')$. From symmetry arguments one can show that they will disappear unless *both* collision partners are polarized. Also, with both collision partners polarized they will be zero if the description of the collision system is chosen "appropriately." Therefore it is important to perform experiments which can supply sufficient information to allow a check on the importance of these coherence terms.

This Letter presents an experimental method which allows the determination of the complete set of eight independent parameters that a full exploitation of the collision geometry may yield. The approach, in which the polarizations of the two excited atoms can be varied independently, gives a direct measure of the influence of coherence and has a more general applicability than to the special case considered here. We give results for two coherence terms for which the detection scheme is simple and, by comparison with cross sections reported earlier,[7] show that coherence effects can by no means be neglected in the analysis. A full discussion of the consequences of this finding and how the completeness provides a tool to pinpoint particularly efficient geometrical approaches by diagonalization of the detection matrix involves rather heavy mathematical machinery and will be presented in a forthcoming full paper.[8]

The experimental setup shown in Fig. 1 is basically the same as that described in Ref. 7 but for one important addition. Two counterrunning thermal beams of Na atoms are intersected at nearly right angles (87°) by laser light from a cw dye laser (Spectra Physics 380D), tuned to the $F_l=2\rightarrow F_u=3$ hyperfine component of the $\mathrm{Na}(3s)\,^2S_{1/2}\rightarrow \mathrm{Na}(3p)\,^2P_{3/2}$ transition. All ions created

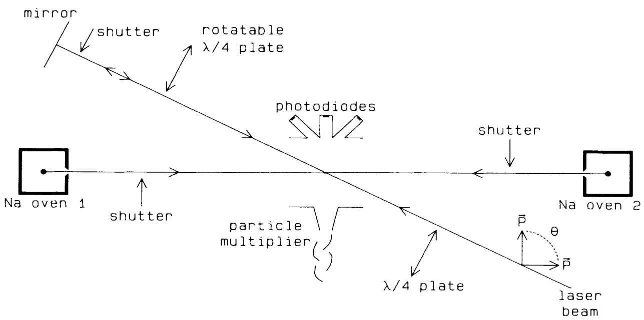
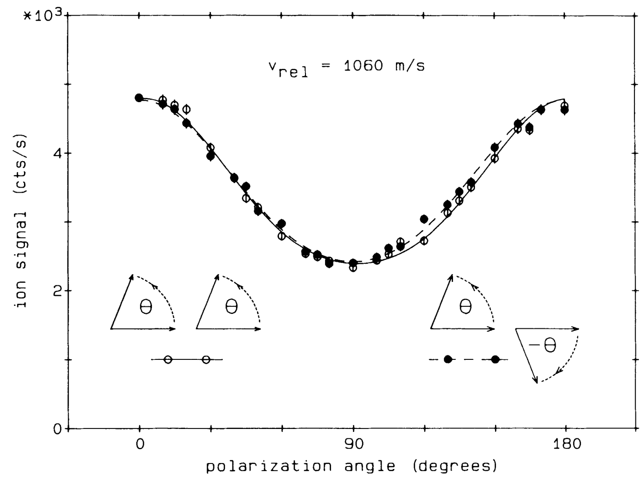
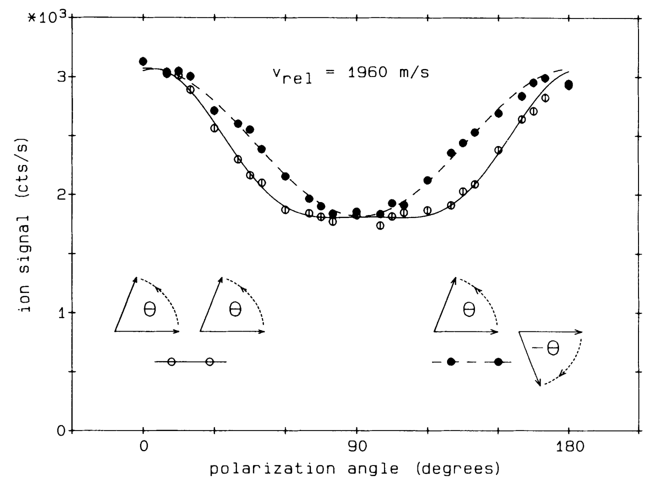



FIG. 1. Schematic view of the experimental setup. For explanation see text.




(a)




(b)

FIG. 2. Polarization dependence measurements with linearly polarized light. Open circles: Both colliding atoms excited with linearly polarized light with the polarization vector at an angle $\theta$ with respect to the collision velocity direction. Filled circles: One atom excited with the polarization vector at $\theta$, and the other at $-\theta$. The lines are least-squares fits by Eq. (3). The difference between the $(\theta,\theta)$ and $(\theta,-\theta)$ cases is $\cos(4\theta)$-like, and depends on collision velocity.

in the interaction region are extracted by a weak electric field, and are counted by a particle multiplier. The fluorescence light is monitored by five photodiodes at various angles. Thereby we are able to monitor the excited-atom density and the atomic polarization. Since the laser intersects the two Na beams at 87°, we can select the velocity class of excited atoms by tuning the laser frequency. The tuned laser frequency is selected and stabilized by means of a third Na beam in a separate vacuum chamber.[9] Moreover, nonperpendicular intersection implies that atoms of beam 2 are only excited by the direct laser beam, whereas atoms of beam 1 are only excited by the reflected laser beam. This allows the polarization of the atoms in beam 1 to be different from those in beam 2, simply by our changing the laser polarization of the reflected beam. To this end we added a rotatable $\lambda/4$ plate, placed in front of the mirror. In this way we did two sets of experiments.

(a) *Linearly ($\pi$) polarized light.*—The polarization vector of the direct beam is at variable angle $\theta$ with respect to the collision velocity direction. The polarization vector of the reflected beam is positioned either at $\theta$ (the main axis of the $\lambda/4$ plate at $\theta$), or at $-\theta$ (the main axis of $\lambda/4$ at 0°). We call these cases $(\theta,\theta)$ and $(\theta,-\theta)$, respectively. As was discussed in Ref. 7 and by Nienhuis,[10] the $\theta$ dependence of the ion production rate can be written as

$$R=R_0+R_1\cos 2\theta+R_2\cos 4\theta, \tag{2}$$

where the Fourier coefficients $R_i$ may be expressed in terms of cross sections and coherence contributions. We use the so-called $L$-picture description, which neglects possible influences of electron spin. Up to now, there is no experimental evidence that this description is incorrect. If we write

$$R^{(\theta,\theta)}=R_0^{(\theta,\theta)}+R_1^{(\theta,\theta)}\cos 2\theta+R_2^{(\theta,\theta)}\cos 4\theta, \tag{3a}$$

$$R^{(\theta,-\theta)}=R_0^{(\theta,-\theta)}+R_1^{(\theta,-\theta)}\cos 2\theta+R_2^{(\theta,-\theta)}\cos 4\theta, \tag{3b}$$

we can derive

$$\begin{aligned} R_0^{(\theta,\pm\theta)} &= \tfrac{1}{1296}(502\bar{\sigma}_{11}+580\sigma_{10}+214\sigma_{00}+27v\mp 36u),\\ R_1^{(\theta,\pm\theta)} &= \tfrac{1}{1296}(-264\bar{\sigma}_{11}+96\sigma_{10}+168\sigma_{00}-36v),\\ R_2^{(\theta,\pm\theta)} &= \tfrac{1}{1296}(18\bar{\sigma}_{11}-36\sigma_{10}+18\sigma_{00}+9v\pm 36u), \end{aligned} \tag{4}$$

in which we use the cross sections $\sigma_{M_1M_2}$ for collisions

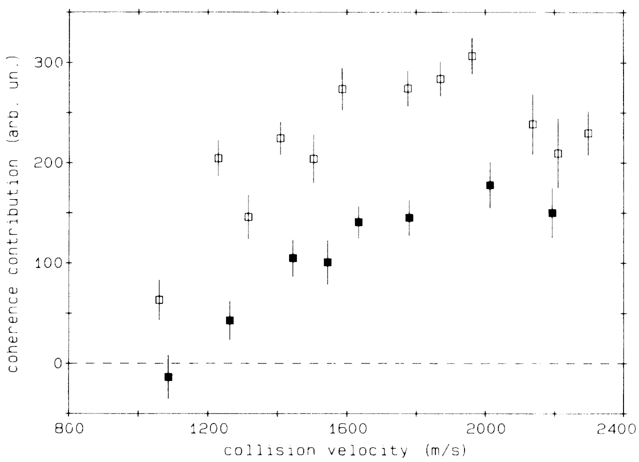



FIG. 3. Results for the coherence term $u$ as a function of collision velocity, for two different series of measurements.

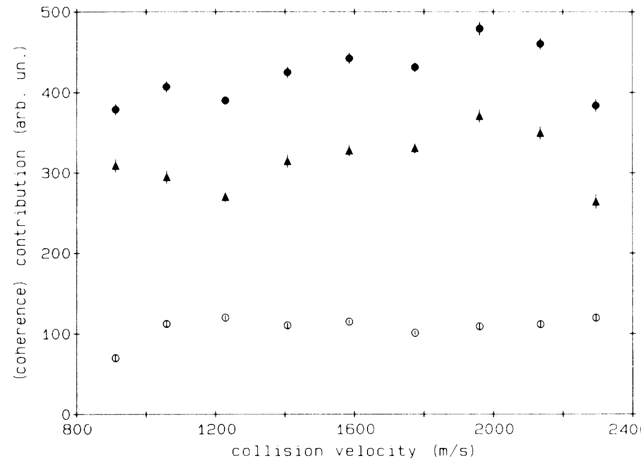



FIG. 4. Measurements with circularly polarized light. Filled circles: Both colliding atoms have parallel angular-momentum projections ($\sigma^{++}$). Open circles: The two atoms have opposite angular-momentum projections ($\sigma^{+-}$). The difference between $\sigma^{+-}$ and $\sigma^{++}$ is equal to the coherence term $r$ (triangles) which gives an important contribution to the ion signal. Vertical axis units are the same as in Fig. 3.

between two atoms in magnetic substates $M_1$ and $M_2$, respectively, quantized along the Na beam direction. Furthermore, $\bar{\sigma}_{11}=\frac{1}{2}(\sigma_{11}+\sigma_{1-1})$, since here we cannot distinguish between $M$ and $-M$. The coherence terms $v$ and $u$ are defined in Ref. 10. In the data analysis, we corrected (4) for non-steady-state conditions.[7] Obviously

$$R^{(\theta,\theta)}-R^{(\theta,-\theta)}=\tfrac{1}{18}u(\cos 4\theta-1). \tag{5}$$

So, by fitting the $(\theta,\theta)$ and $(\theta,-\theta)$ measurements separately with (3a) and (3b), one easily obtains $u$, as well as a check on the experimental conditions, since within experimental error one should find the relations

$$\begin{aligned}&R_0^{(\theta,\theta)}-R_0^{(\theta,-\theta)}=R_2^{(\theta,-\theta)}-R_2^{(\theta,\theta)},\\&R_1^{(\theta,\theta)}=R_1^{(\theta,-\theta)}.\end{aligned} \tag{6}$$

We measured the $\theta$ $(-\theta)$ dependent ionization signals in two series at various collision velocities. Some results are shown in Fig. 2. The two curves should be identical if the coherence term is zero. Clearly it is not. At all velocities we found the relations (6) to be fulfilled within the experimental error. The strong velocity dependence of the coherence contribution $u$ is shown in Fig. 3. Considering the fact that $u$ is a small difference between two large numbers, the discrepancy between the two different measurement series may be caused by, e.g., a small misalignment of the $\lambda/4$ plate. However, both measurements clearly have the same trend.

(b) *Circularly polarized light.*— By use of another $\lambda/4$ plate the direct beam becomes circularly ($\sigma^+$) polarized. The $\lambda/4$ plate in the front of the mirror is either out of the beam (then the photons of the reflected beam have not changed angular momentum with respect to the collision frame), or in the beam with its main axis at an arbitrary angle (reflected-beam photons now have their angular momentum pointing in the opposite direction). In this case steady-state conditions are fulfilled, because there is no interference with other nearby hyperfine transitions.[11] Now we can derive

$$R^{+\pm}=\tfrac{1}{4}\bar{\sigma}_{11}+\tfrac{1}{2}\sigma_{10}+\tfrac{1}{4}\sigma_{00}+\tfrac{1}{8}v\mp\tfrac{1}{2}r, \tag{7}$$

i.e.,

$$R^{++}-R^{+-}=-r. \tag{8}$$

The results for the measurements of $R^{++}$ and $R^{+-}$ are shown in Fig. 4. During the measurements steady-state conditions were checked by use of the fluorescence signals of the various photodiodes at different angles. The coherence contribution $r$ is large and almost independent of collision velocity. This measurement showed very good reproducibility.

The measured coherence contributions $u$ and $r$ are comparable in size to the cross sections presented in Ref. 7 (of which the vertical axis is in the same units), and so it is in general not justified to assume vanishing coherence contributions (which, in fact, was done in Ref. 7 to get the cross sections).

The experimental geometry allows extraction of up to seven independent parameters. In the $L$ picture, there are four cross sections $\sigma_{M_1M_2}$ (two of which we average), and four coherence terms. Two of the coherence terms can be determined in a direct way, as presented here. For the two other coherence terms and the cross sections, more complicated detection schemes are needed. However, we have succeeded in determining these terms too, and thus the full set of cross sections and coherence terms. These results, as well as a full discussion of theory and experiment, will appear in a forthcoming paper.[8]

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