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THE "COMPLETE" p-p EXPERIMENT

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We are reporting on an experiment leading from an initial optically pumped p to a final p state, in which all relevant amplitudes and phases can be determined. In particular, the charge exchange reaction $H^+ + Na(3p_{\pm 1}) \rightarrow H(2p) + Na^+$ was studied at a projectile energy of 2 keV. This is the first quantum mechanically complete experiment with a non-isotropic initial state leading to a non-isotropic final state.

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1. *Introduction*

Optically pumped, state-prepared atoms have been used extensively in atomic collision physics, and a wide variety of processes have been studied. Such systems can be adjusted with great flexibility to the special requirements of investigated phenomena. For example, laser-prepared collision partners can carry initial alignment and orientation and thus illuminate the role of "shape" and "rotation" of the initial atomic state.

Mainly due to its easy handling, as well as to the availability of the required laser radiation, collision studies employing a Na target are at present at the centre of attention. In particular, charge exchange in H^++Na collision reaction around 1 keV [1–13] is serving as a test case in which a rather complete understanding is reached on the basis of large-scale *ab initio* computer calculations. Several aspects of its collision dynamics have been studied: total [1–6] and angle-differential cross-sections [7–9] , alignment effects [8,10,11], the so-called "left-right asymmetry" [8,12,13] which is related to collisionally accumulated phases of the involved electron states, and other "propensities" [14]. This system, also being of some technical interest, provides a convenient model for the study of electron dynamics in ion-atom interactions; similarly to other few-electron systems [15], it may even provide a ground for the most severe ("quantum-mechanically complete") test of our understanding of the collision. For such an experiment, preparation of the initial state (e.g., Na($3p_m$) as well as identification of the final channel (e.g.,H($2p_m$)) is essential since knowledge about all states is required. Our aim is to perform a "complete" experiment involving the anisotropic 3p initial state. The process under investigation is:

$$\mathrm{H}^+ + \mathrm{Na(3s,3p)} \to \mathrm{H(2p)} + \mathrm{Na}^+. \qquad (1)$$

While for the coherent "s-p excitation" case [16], the process is completely characterized by three parameters, the "p-p" case is considerably more complex. Three initial and three final magnetic substates generally demand nine complex amplitudes (Fig. 1). Symmetry considerations reduce the 17 real quantities to nine: five amplitudes and four phases are required (including the phase between the initial $m = 1$ and $m = 0$ states) [17].

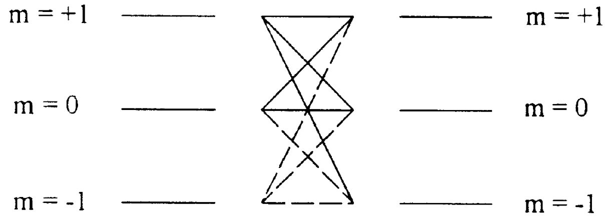

Fig. 1. Schematic representation of the various excitation pathways for $\mathrm{p}_{0,\pm1} \to \mathrm{p}_{0,\pm1}$ *reactions. Dashed lines indicate amplitudes which are redundant due to the reflection symmetry.*

At least nine independent measurement are needed for a quantum mechanically complete description of the p-p charge exchange process. A proper choice of the experimental geometry, i.e., high symmetry as well as a pure initial state, simplify the analysis and make the complete determination possible. A preferable condition is, e.g., pumping with circularly polarized light which propagates collinearly with the ion beam. In this case a pure $m = 1$ (or $m = -1$) state is prepared in the photon frame, which is in this case identical to the collision frame. Other laser pumping geometries prepare an incoherent (pumping with linearly polarized light) or coherent (pumping with circularly polarized light) superposition of substates. Due to the Na hyperfine interaction, no geometry allows to prepare a pure $m = 0$ state.

2. *Experimental method*

The experimental set-up is shown in Fig. 2. A proton beam from a duoplasmatron ion source is accelerated to an energy of 2 keV. After collimation to better than $0.03°$

in a differentially pumped drift tube, the ion beam is injected into the target chamber. The background pressure is 1×10^{-7} mbar. The target chamber is free of electric fields to avoid quenching of collision-produced H(2s), and the earth's magnetic field is compensated by Helmholtz coils to values smaller than 5 μT. About 25 mm before the interaction region, the ion beam is cleaned from neutrals with 3 pairs of deflector plates. A beam from a single mode Ar ion/dye laser combination (Coherent) tuned to the Na(3s)^{2}S$_{1/2}$($\overline{F} = 2$) $\rightarrow$Na(3p)^{2}P$_{1/2}$($F = 3$) transition (589 nm) counterpropagates to the ion beam. In the interaction region, the ion and the laser beam are crossed at right angles by a Na-atom beam. The sodium beam is produced in a two-stage oven which reduces the dimer content to less than 1%. After skimming, the beam is collimated to a divergence of less than $0.2°$. The Na-atom density in the collision area is 10^9 to 10^{10}cm^{-3} ; it is kept constant by controlling the oven temperature. Lyman-α radiation (121.6 nm) emitted from

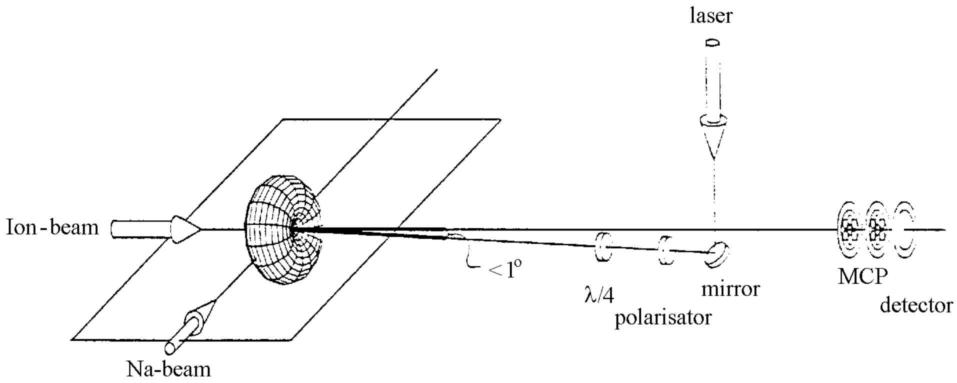

Fig. 2. Experimental set-up (schematic).

capture-produced H(2p) is detected in a solar-blind photomultiplier. It has a MgF$_2$ window, providing a sensitivity region between approximately 115 and 200 nm; the specified quantum efficiency for Lyman-α radiation is about 15%. The photomultiplier is placed at approximately the "magic angle" ($\vartheta_\gamma = 54.7°$ relative to the ion beam axis), in the plane defined by the atomic and the ion beam; it views a length of ion beam around the interaction region of about 6 mm, thereby subtending a solid angle of approximately 0.3 sr. The Lyman-α photons are measured in coincidence with neutralized projectiles which are scattered through defined angles into a position-sensitive detector. This detector consists of 2 micro-channel plates in a chevron set up and a divided-anode array which is composed of 16 individual electrodes covering 4 azimuthal angles $\varphi_s(0, \pi/2, \pi, 3\pi/2)$ for each of four centre-of-mass scattering angles ϑ_s. The arrangement allows to simultaneously and continuously monitor the proper alignment of the primary beam. By choosing different distances (2.0, 2.6 and 3.6 m, respectively) between the collision region and the particle detector, scattering angles between $0.05°$ and $0.4°$ can be covered. The relative efficiency for particle detection in each individual anode segment is determined in a separate experiment. To this end, the proton beam is scattered on a dense N$_2$ target instead of the Na target; this gives a rather homogeneous scattering profile within the angular range of interest. The efficiency determination is repeated after every few runs.

Pulses from the 16 anodes are processed separately; particular care has been taken to eliminate cross-talk. The pulses serve as stop input for 16 time-to-digital converters; the photomultiplier gives the necessary start signal. The time resolution (about 15 ns) is limited by the finite length of ion beam viewed by the photon detector. Data processing is performed by a personal computer which also controls the experiment. The Na-beam is mechanically chopped and the signals detected in synchronisation with the chopper. Data are taken under alternating Na beam-on and beam-off as well as laser-on and laser-off conditions. All the data are taken under single-collision conditions with proper correction for background contributions as obtained from Na beam-on and beam-off measurements. In the case of "laser-on", both species, sodium in the $3S_{1/2}$ and the $3P_{3/2}$ state are present. We are assuming that the pumping mechanism prepares a statistical mixture, rather than a coherent superposition of atoms in the ground and excited states. Fractions of target atoms in the excited state can be determined through the momentum transfer per absorbed photon, h/λ, which results in a recoil of the sodium atoms, as described in Ref. 5. A small fraction of the laser beam is used for frequency stabilisation.

3. Data evaluation and results

The experiment is based on the coincident detection of scattered H atoms and emitted Lyman-α radiation. Detection of photon and of scattered H atom is direction-sensitive according to polar $(\vartheta_\gamma, \vartheta_s)$ and azimuthal $(\varphi_\gamma, \varphi_s)$ angles; the coincidence requirement identifies the scattering plane and the photon direction for each interaction event. A typical time spectrum obtained from one detector segment is shown in Fig. 3. The peak at about 4.2 μs contains scattering events of 2 keV H^+ projectiles; the one at about 5.8 μs is caused by 2 keV H_2^+ projectiles, also contained in the ion beam. The time spectrum allows to separate the contributions from both components.

The coincidence requirement strongly discriminates against capture from background gas. The number of true coincidences is obtained in the usual way by subtracting the random coincidences from the total coincidences within a given time window around the "true" peak. The coincident light-emission intensity for the general case of a p-p scattered particle-photon case is given by [17]:

$$I = \sum_{K=0,2} \left\{ B_K \bar{G}_K \sum_{Q=0}^{K} (2-\delta_{Q0}) d(\vartheta_\gamma)^K_{0Q} \right. \tag{2}$$

$$\left. \times \sum_{K_0 Q_0} Re[< T(L_0)^+_{K_0 Q_0} \times t_{00} > A(KQ, K_0 Q_0) e^{i[Q(\varphi_\gamma - \varphi_s) + Q_0 \varphi_s]}] \right\}$$

$$+ \sum_{K=0,2} \left\{ \{ B_K \cdot g_{11K} \sum_{Q=0}^{K} (2-\delta_{Q0}) d(\vartheta_\gamma)^{(K)}_{0Q} \sum_{Q'=-1}^{1} (1Q', 1Q-Q'|KQ) \right.$$

$$\times \sum_{K_0 Q_0} Re[< T(L_0)^{+}_{K_0 Q_0} \times t_{1,Q-Q'} > A(KQ', K_0 Q_0) e^{i[Q\varphi_\gamma + (Q_0 - Q')\varphi_s]}] \Bigg\} .$$

$d(\vartheta_\gamma)_{KQ}$ are the elements of the rotation matrix which transform the photon detec-

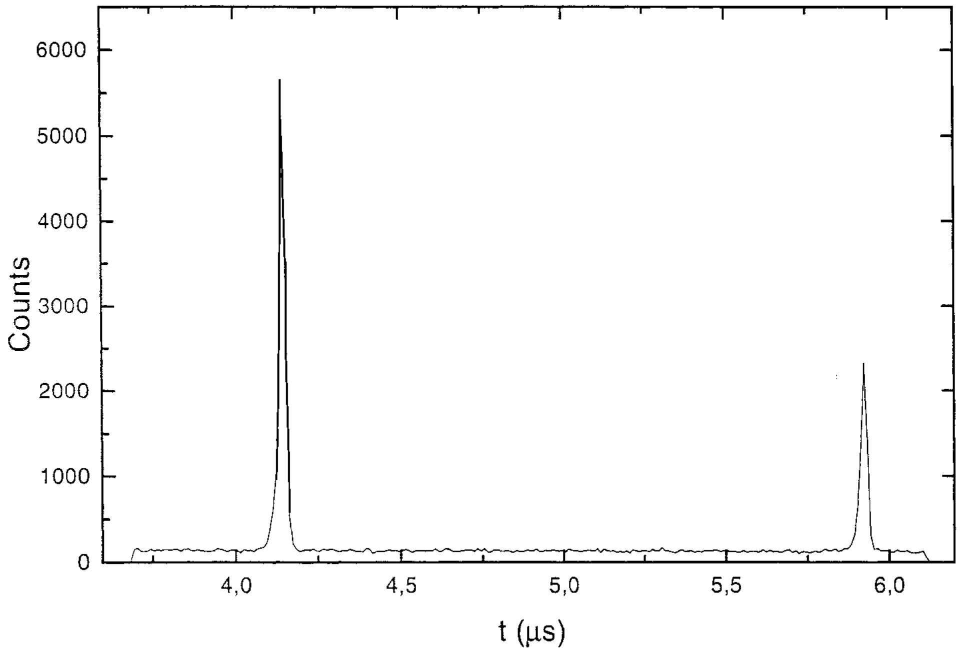

Fig. 3. A typical Lyman-α/scattered H *particles coincidence time spectrum at the scattering angle of* $0.069°$ *in the centre-of-mass system.*

tor (located at $\vartheta_\gamma, \varphi_\gamma$) into the frame in which the final state multipoles are defined, and $G^{Q' q_0 Q}_{K' k_0 K}$ are generalized perturbation coefficients given by the expression

$$G^{Q' q_0 Q}_{K' k_0 K} = (K'Q'k_0 q_0 | KQ) \sum_J (2J+1)^2 \tag{3}$$

$$\times \sqrt{(2K'+1)(2k_0+1)(2K+1)} \left\{ \begin{array}{ccc} K & O & K \\ L & \frac{1}{2} & J \\ L & \frac{1}{2} & J \end{array} \right\} \left\{ \begin{array}{ccc} K & k_0 & K \\ L & \frac{1}{2} & J \\ L & \frac{1}{2} & J \end{array} \right\}$$

$$\equiv (K'Q'k_0 q_0 | KQ) g_{K' k_0 K} .$$

$(|)$ denote Clebsch-Gordan coefficients and $\{--\}$ standard $9j$-symbols. The quantities $A(KQ, K_0Q_0)$ describe the scattering dynamics; they are defined as

$$A(KQ, K_0Q_0) = \sum_{\substack{MM'\\ M_0M_0'}} (-1)^{L_0-M_0+L-M} \tag{4}$$

$$\times (LM', L-M|KQ)(L_0M_0', L_0-M_0|K_0Q_0)f(M'M_0')f(MM_0)^*,$$

and $f(MM_0)$ are the corresponding amplitudes for scattering from an initial M_0 to a final M state, after separation of the spin dependence. It is assumed that the total spin is conserved during the collision so that the spin component of the active electron *immediately after the collision* is the same as that in the initial state, and the hyperfine interaction in the final state is negligible.

The goal of the "complete experiment" is the determination of all amplitudes $f(MM_0)$ and the corresponding phases, contained in $A(KQ, K_0Q_0)$. We point out that in contrast to the case of an isotropic initial state [15], the presence of the polarized spin causes the expression for the angular distribution of emitted radiation to contain all state multipoles of rank 0,1 and 2. To obtain the sign of the phase, it is therefore sufficient to perform a correlation measurement; a polarization (Stokes parameter) analysis of the final state is not necessary.

The complexity of the relation (2) suggests the use of geometries of high symmetry and, if possible, a pure initial state. Our experimental geometry allows preparation of pure $m = 1$ (or $m = -1$) initial states, and the photon detector is placed at the "magic angle" $\vartheta_\gamma = 54.7°(i.e., 3cos^2\vartheta_\gamma - 1 = 0)$. In this case relation (2) simplifies to [18]:

$$\begin{aligned} I(\vartheta) &\approx 3[f(-11)^2 + f(01)^2 + f(11)^2] \\ &+2Re[f(01)f(11)^*]\cos(\varphi_s) + 2Im[f(01)f(11^*)]\sin(\varphi_s) \\ &+2Re[f(-11)f(11)^*]\cos(2\varphi_s) + 2Im[f(-11)f(11)^*]\sin(2\varphi_s). \end{aligned} \tag{5}$$

Thus, seven independent measurements are required for a complete determination of the charge exchange process involving the $3p, m = \pm 1$ state. As mentioned above, our position sensitive particle detector consists of 16 (ϑ_s, φ_s) independent segments, placed at four azimuthal angles 0, 90, 180, and 270 degrees for each of the four scattering angles ϑ_s. We were performing 16 independent measurements in one run; some azimuthal angles are redundant. From the measured intensities, we extract the required quantities for every scattering angle by use of the following relations:

$$\begin{aligned} \sum |fi|^2 &\approx \frac{1}{12}[I(0°) + I(90°) + I(180°) + I(270°)] \equiv \sigma \\ Re\,[f_{01}\ f_{11}^*] &\approx \frac{1}{4}[I(0°) - I(180°)] \\ Im\,[f_{01}\ f_{11}^*] &\approx \frac{1}{4}[I(90°) - I(270°)] \end{aligned} \tag{6}$$

$$Re\,[f_{-11}\, f^{*}_{11}] \;\;\approx\;\; \frac{1}{4}\,[I(0^\circ) + I(180^\circ) - I(90^\circ) - I(270^\circ)]\,.$$

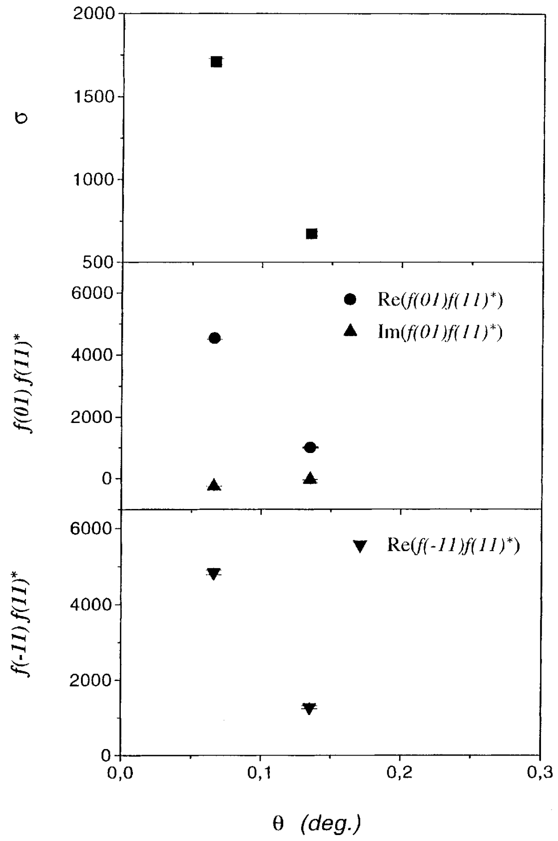

Fig. 4. The $3p, m = 1 \rightarrow 2p$ charge exchange reaction: a) scattering angle-dependent differential cross-section; b) Real and imaginary part of the phase $f(01)f^*(11)$ c) Real part of the phase $f(-11)f^*(11)$.

The finite acceptance ranges of the various angles involved have been included in the following way: (i) The effect of the finite photon detector acceptance angle $(54.7^\circ \pm 9^\circ)$ is small and has been neglected; (ii) the effect of the finite azimuthal acceptance (approximately $\pm 15^\circ$) has been taken into account by an approximate integration over the corresponding range in Eq. (5). Although the Lyman-α coincidence requirement selects population of the H(2p) final state, this does not entirely exclude cascade transitions into this level. However, due to the finite length of the particle beam viewed by the photon detector, only short-lived higher n states can contribute to the coincidence signal; therefore, they have been neglected.

In Fig. 4a we show the scattering-angle-dependent cross-sections σ for electron capture from excited Na($3p_{\pm1}$) state into H(2p) final state. So far, no theoretical calculations are available for comparison. Figures 4b and c show the real and the imaginary part of the phase $f(01)f^*(11)$ and the real part of the phase $f(-11)f^*(11)$. By use of the definition of the quantum mechanical phase, the sin and cos part of the phase can be calculated from the measured data. We found that the phase angle between the phases f_{01} and f_{11} is close to zero at the measured scattering angles. Since the scattering process is strongly forward peaked, the detectors placed at larger scattering angles detected very weak signal. Evidently future measurements should preferrably be performed at somewhat smaller impact energies, in order to obtain appreciable contribution at the larger scattering angles. Measurements with a projectile energy of 1 keV are in progress.

4. Summary

A collision leading from an initial p to a final p state is, in general, characterized by nine real amplitudes and phases. As an example, collinear laser pumping of Na(3s) with circularly polarized NaD light produces a spin-polarized pure Na(3p,$m = \pm1$) state; the "theory of the experiment" shows that the relevant amplitudes and phases can be determined by means of a simple angular correlation (photon-scattered particle coincidence) experiment instead of a Stokes parameter measurement. We have performed the first experiment which will allow a complete determination of all amplitudes and phases, and present preliminary experimental results for the process Na($3p_1$)+H^+ $\rightarrow$ H(2p)+Na^+ at 2 keV.

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"CJELOVITO" p-p MJERENJE

Načinili smo mjerenje procesa u kojem početno optički pumpano stanje p prelazi u konačno stanje p uz određivanje svih relevantnih amplituda i faza. Posebice, proučavala se reakcija izmjene naboja $H^+ + Na(3p_{\pm 1}) \rightarrow H(2p) + Na^+$ pri energiji protona od 2 keV. Ovo mjerenje predstavlja prvi kvantno-mehanički cjelovit eksperiment u kojem se mjerio prijelaz neizotropnog početnog stanja u neizotropno konačno stanje.