Preparation of Molecular States in an Ion-Atom Crossed-Beam Scattering Experiment by Laser Excitation

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In Na^+-Na* scattering in the energy range $E_{c.m.}$ = 25–50 eV the molecular states σ, π^+, and π^- of the resulting ionic quasimolecule Na_2^+ are prepared by laser-optical pumping of the sodium atom to the $3^2P_{3/2}$, $F = 3$ state. Selection is made with linearly or circularly polarized light. With this novel approach, one can discuss the transition from the space-fixed to the body-fixed reference frame in detail. There is clear evidence for conservation of reflection symmetry with respect to the scattering plane and the implications are considered.

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The detailed investigation of heavy-particle collisions by analysis of the light emitted after the collisional excitation of atoms has made progress in the last years. From a measurement of direction and polarization of the emitted photons in coincidence with the scattered ions one may obtain—in principle—population amplitudes and phase differences of the magnetic sublevels of the excited atom. For collision energies in the kiloelectronvolt energy range measurements were successfully done for the systems[1-3] He^+ on He and Be^+ on He, Ne, and Ar and in the region of some hundred electron volts for K on He, Ne, and Ar.

In this work the quasi one-electron system Na^+-Na and the very low energy range were chosen to get a "pure case," that is to say in the experiment the state of the system before and after the collision is known accurately. We are able to control the translational momentum as well as the internal energy of the collision partners. The aim of the present scattering experiment is to prepare asymptotically defined atomic states and study their development through the molecular states. The sodium target atoms are aligned or oriented by optical pumping with polarized laser light. In this experiment we measure for the first time a complete set of atomic alignment and orientation parameters[4] for ion-atom scattering with this powerful technique, which has been applied before to electron-atom scattering.[5] Figure 1 shows a schematic picture of the scattering apparatus.

An energy-loss spectrum for the scattering of Na^+ from Na at $E_{lab} = 95$ eV and $\theta_{lab} = 2°$ is shown in Fig. 2. Without light one sees the inelastic scattering process

$$Na^+ + Na(3^2S) + \Delta E_{kin} \rightarrow Na^+ + Na^*(3^2P). \qquad (1)$$

Differential cross sections for process (1) have been measured at $E_{c.m.} = 37.5$–57.25 eV by Hormes[6] and the influence of the inelastic channel on the differential cross section for charge transfer in the energy range $E_{lab} = 250$–1250 eV has been studied in an experiment by Wijnaendts van Resandt *et al.*[7] With the laser on one observes a decrease of the inelastic signal in process (1), which leads to the conclusion that approximately 12% of the sodium atoms are pumped to the excited $3^2P_{3/2}$ state. On the high-energy side one recognizes the superelastic scattering process

$$Na^+ + Na^*(3^2P_{3/2}, F = 3) \rightarrow Na(3^2S) + Na^+ + \Delta E_{kin}. \qquad (2)$$

Figure 3 shows the angular dependence of the inelastic and superelastic processes at $E_{c.m.} = 47.5$ eV with a pronounced maximum at $\theta_{c.m.} = 4°$. This structure results from rotational coupling at the crossing between the $^2\Sigma_u^+$ ground and $^2\Pi_u^+$ excited

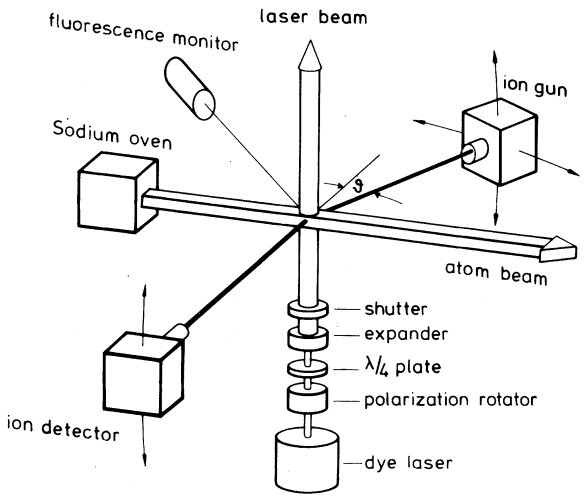

FIG. 1. Schematic diagram of the apparatus.

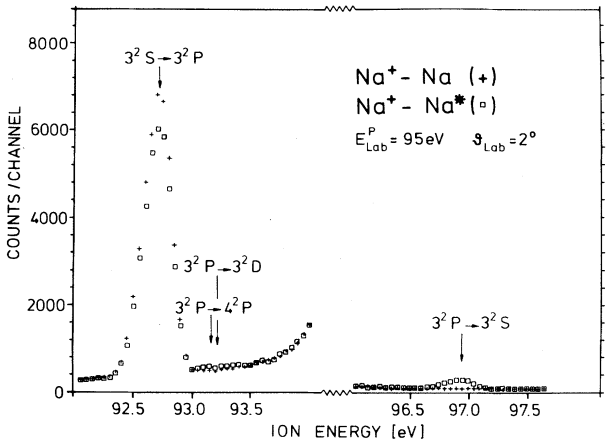

FIG. 2. Energy-loss spectrum for $Na^+ + Na^*$ ($3^2P_{3/2}$) and $Na^+ + Na$ (3^2S) scattering. Note that the elastic peak is suppressed.

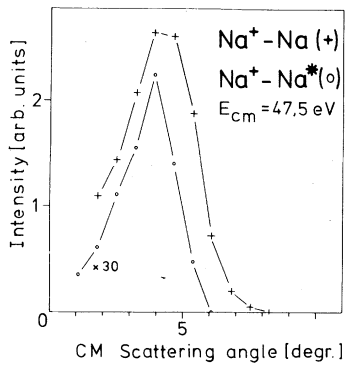

FIG. 3. Angular dependence for $Na^+ + Na^*$ ($3^2P_{3/2}$) and $Na^+ + Na$ (3^2S) scattering.

states of Na_2^+ at $R \sim 5$ a.u., which can be seen from the calculated potential energy curves.[8,9] For rotational coupling the transition probability is highest when the classical turning point is near the crossing. Thus one has a small inelastic and superelastic cross section for impact parameters (i.e., scattering angles) differing from those which lead to a turning point near $R \sim 5$ a.u. From Fig. 3 one sees also that this rotational coupling is the only important mechanism in this energy and angular range for the processes (1) and (2), since there is no detectable superelastic- or inelastic-scattering signal for larger scattering angles.

The experimental results presented above show that for the model system Na_2^+ and in the energy range we have chosen, one obtains a clear picture of the "conventional" part of the scattering process. On the basis of this we now want to discuss the goal of our following experiment, the measurements of the dependence of the superelastic scattering signal on alignment and orientation of the excited sodium atom. Figure 4 shows in the upper half the scattering intensity for linearly polarized laser light directed perpendicular to the scattering plane, as a function of the angle θ_E between the direction of the incoming ion and the electric vector of the laser light. For $\theta_E \sim 90°$ the π^+ molecular state is preferentially prepared, which gives clearly more scattering signal than preparing the σ state at $\theta_E \sim 0°$. The assignments σ and π^+ refer to the configuration at infinite internuclear distances of Na* and Na^+. From the fact that we reach the $^2\Pi_u^+ - ^2\Sigma_u^+$ crossing via the $^2\Pi_u^+$ potential curve and because the asymptotically prepared π^+ molecular state gives the strongest deexcitation signal we conclude that the molecular picture is valid from large Na^*-Na^+ distances on. Looking at the intensities in Fig. 4 one must keep in mind that we are preparing here aligned atoms. We have an incoherent su-

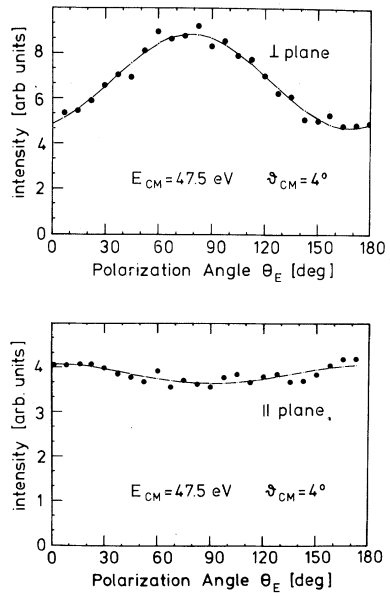

FIG. 4. Dependence of the superelastic signal in $Na^+ + Na^*$ ($3^2P_{3/2}$) → $Na^+ + Na$ (3^2S) + ΔE_{kin} scattering on polarization angle of linearly polarized laser light, crossing the scattering zone at right angle with respect to the scattering plane (⊥) as shown in the upper half or propagating in the scattering plane (II) as shown in the lower half. The scattering plane is defined by the directions of the incoming and outgoing ion. In the measurement ⊥ plane $\theta_E = 80°$ refers to preparing a π^+ molecular state and $\theta_E = 170°$ to preparing a σ state. In the measurement II plane $\theta_E = 0°$ refers to preparing a σ molecular state and $\theta_E = 90°$ to preparing a π^- state. The thin line is a fit to the measured data points.

perposition of Na* substates[10] with the ratio $p_z:p_x:p_y = 2.5:1:1$, where the z axis is in the direction of the electric vector of the laser light and $p_{x,y,z}$ refer to the population of the three orthogonal $3p$ orbitals. Upon closer inspection of the superelastic-scattering signal the maximum is found for $\theta_E^{\max} = \gamma = 80° \pm 5°$. The slight deviation of the maximum from 90° results from the merging of atomic and molecular pictures up to a finite internuclear distance R_1, where the electron orbital is finally locked to the internuclear axis. By a comparison of the energy difference of the Π and Σ potentials[9] at large internuclear distances with the energy of the rotation of the p orbital in the space-fixed frame one finds, for an impact parameter $b = 5$ a.u., $\theta_E = 89.2°$ and $R_1 \sim 375$ a.u. which does not agree with our measurements. For the case of these long-range potentials with R^{-3} dependence one needs a dynamical treatment. Dynamical semiclassical calculations have been done by Grosser,[11] who neglects, however, the impact parameter dependence of the merging radius. We obtain from his formula $\theta_E = 82°$ and $R_1 = 35$ a.u. which agrees well with our results. In thermal collision processes[12] Na*-N_2 where impact parameter and locking radius R_1 are almost equal the Grosser formula is not applicable; it holds only for $b/R_1 \ll 1$. Figure 4 shows in the lower half the polarization dependence for the laser light propagating in the scattering plane. In this case σ ($\theta_E = 0°$) and π^- ($\theta_E = 90°$) molecular states are preferentially selected. For preparation of a π^- state the $\vec{E}$ vector of the light is perpendicular to the scattering plane. The measured collision deexcitation probabilities resulting from the different molecular states are $P_{\pi^+} = 0.7 \pm 0.1$, $P_{\pi^-} = 0.1 \pm 0.1$, and $P_\sigma = 0.2 \pm 0.1$. The outcome of the measurements shown in Fig. 4 is that reflection symmetry with respect to the scattering plane is conserved within our error limits. This is a result of our measurements and not assumed as it is often done for the case of $S \rightarrow P$ transitions. The value of $P_{\pi^-} = 0.1 \pm 0.1$ shows some indication for symmetry-changing (i.e., spin-flip) processes, which may arise because at our energies the collision time is on the order of the precession time of the electron spin.

We prepare oriented excited sodium atoms with circularly polarized light [right-hand circular (RHC) or left-hand circular (LHC)] directed perpendicular to the scattering plane. The measured circular asymmetry $P_3 = (I_{\mathrm{RHC}} - I_{\mathrm{LHC}})/(I_{\mathrm{RHC}} + I_{\mathrm{LHC}})$ is $P_3 = 0.28$ for $E_{\mathrm{c.m.}} = 47.5$ eV and $P_3 = 0.24$ for $E_{\mathrm{c.m.}} = 25$ eV at $\tau = 180$ eV deg, where $I_{\mathrm{RHC,LHC}}$ are intensities of the scattered ions. This result is explained by understanding the dynamics in the transition region from the space-fixed to the body-fixed description. As has been pointed out by Grosser[11] the merging of the atomic (space-fixed) frame and the molecular (body-fixed) frame gives rise to Σ-Π transitions, where the atom remains in the 3^2P excited state. This we saw before since $P_\sigma \neq 0$. With use of RHC or LHC polarized light all excited σ and π molecular states are simultaneously and coherently populated with a phase difference of $\Delta\varphi = \pm 90°$. The difference in the scattering signal for RHC and LHC light is a result of the different interference patterns of the particle waves. These experience the Σ and Π potentials at large internuclear distances and in the merging region Σ-Π transitions take place and the waves interfere on the way through the exit of the $^2\Pi_u^+$-$^2\Sigma_u^+$ crossing. The measurements with circularly polarized light are extremely sensitive to the Na^+-Na potential at large internuclear distances. The situation cannot be described by simply assuming a R^{-3} dependence of the interaction potential, because the *gerade-ungerade* potentials[8] differ considerably in that range. The results of our measurements will be presented more completely in form of the density matrix in a forthcoming publication.[13] In this paper we hope, however, to have given a qualitative insight into the dynamics of the forming of a molecule in a collision process.

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Role of Damping and *T* Invariance in Induced Transitions in H(2*S*)

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An interference term has been observed in an induced transition between damped substates of $2S_{1/2}$ atomic hydrogen which is formally odd under time reversal. The magnetic-field dependence of this term through the $2S_{1/2}$-$2P_{1/2}$ level crossing provides a distinct characterization of the interaction of the atom and the radiation field. The results are central to the design and interpretation of the hydrogen parity experiment.

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There is a considerable literature concerning the behavior of unstable particles in external fields.[1] Of recent interest has been the 2*S* state of hydrogen and hydrogenic ions in the context of investigations of parity nonconservation[2,3] and of the diverse interference phenomena in the angular distribution of Lyman-α radiation from quenching in electric fields.[4] We present here the results of a novel experiment with atomic hydrogen bearing on the role of time-reversal (T) symmetry in induced transitions between damped $2S_{1/2}$ substates.

The initial motivation for this work was to demonstrate experimentally the validity of the formulation of our experiment to investigate parity-nonconserving (PNC) weak interactions in atomic hydrogen.[5] This experiment is based on observation of a pseudoscalar interference term in a suppressed microwave electric dipole transition between the (initial) α_0 and (final) β_0 substates[6] of the metastable $2S_{1/2}$ state in a dc magnetic field, $\vec{B}$, set near the crossings of the β substates and the substates e of the strongly damped $2P_{1/2}$ state (Fig. 1). The transition is induced by mixing of β_0 and e_{+1} with a small applied dc electric field, $\vec{E}$, perpendicular to $\vec{B}$, and of β_0 and e_0 by the putative PNC interaction. The microwave electric field, $\vec{\epsilon}$, and $\vec{E}$ and $\vec{B}$ are arranged in a configuration which breaks inversion symmetry (Fig. 2 with $\psi = 0$). As a consequence

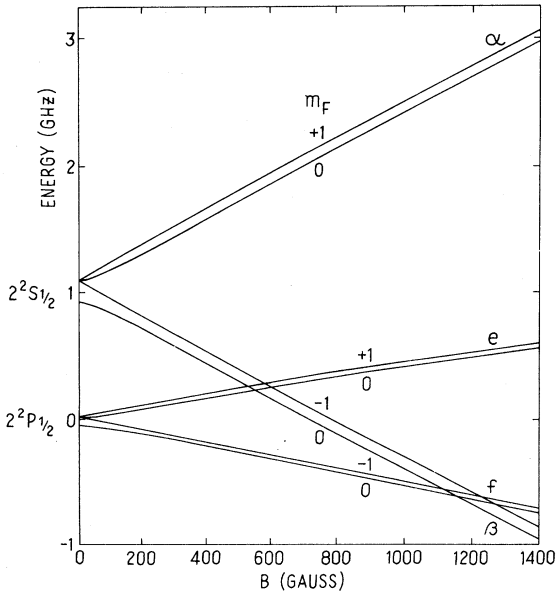

FIG. 1. Zeeman diagram of the $2S_{1/2}$ and $2P_{1/2}$ states of hydrogen.

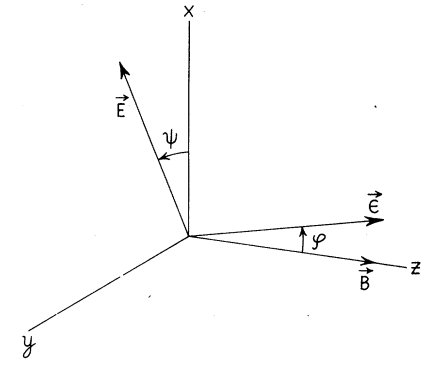

FIG. 2. Field configuration in the transition region.