# State-changing in Na($ns,np$)-Xe collisions

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In thermal-energy collisions between sodium atoms in $ns$ or $np$ states ($32 \leq n \leq 52$) and xenon atoms, depopulation of the Rydberg states is observed to occur predominantly through state-changing reactions of the type Na($ns$ or $np$) + Xe → Na($n-1$, $l \geq 2$) + Xe. For the present range of $n$, the measured cross sections increase with $n$ and lie between 1500 and 2600 Å$^2$ for $ns$ states and between 2000 and 4200 Å$^2$ for $np$ states. Implications of the data are discussed.

In recent years a number of studies of state changing in thermal-energy collisions between alkali atoms in high Rydberg states and the rare gases have been reported.[1-3] In the case of sodium much of this work was focused on collisions involving $d$ or $f$ states. However, collisional depopulation of Na($ns$) states by helium, argon, and xenon has been investigated by Gallagher and Cooke[4] for states with principal quantum numbers $n$ in the range 6 to 11. In the present work we have investigated depopulation of $s$ and $p$ states in collisions with xenon for values of $n$ in the range 32 to 52.

The experimental technique and apparatus have been described in detail elsewhere.[5] Sodium atoms in a thermal beam are excited, in the presence of xenon, to high Rydberg states by pulsed-laser-induced photoexcitation. The $p$ states are produced by direct excitation from the ground state, and the $s$ states by two-step excitation via the intermediate $3^2P_{1/2}$ state. Following excitation the atoms are allowed to interact with the target gas for a selected time $t$, within the range 0–7 $\mu$sec, after which the remaining parent and product Rydberg atoms are ionized in a time-dependent electric field. Since Rydberg atoms in different quantum states ionize at different field strengths, measurement of the field dependence of the ionization signal permits identification of the excited atoms present in the interaction region. This technique, referred to as selective field ionization (SFI), has been widely used to analyze excited-state distributions.[5-7] The SFI data obtained here show that state changing occurs predominantly through reactions of the type

$$\text{Na}(ns \text{ or } np) + \text{Xe} \rightarrow \text{Na}(n-1,\ l \geq 2) + \text{Xe} \qquad (1)$$

and that collisions populate states having a range of values of $l$ and $|m_l|$. In addition, no significant collisional ionization is detected.

Absolute rate constants for state changing are obtained by measuring, using SFI, the time dependence of the parent state population $N(t)$ and the state-changed population, viewed as a reservoir of population $R(t)$. The time dependence of $R(t)$ is given by

$$\frac{dR(t)}{dt} = \rho k N(t) - bR(t) \ , \qquad (2)$$

where $\rho$ is the xenon density, $k$ the rate constant for state-changing collisions, and $b$ the reservoir decay rate. $k$ is determined by fitting the integral form of Eq. (2) to the data. Cross sections are obtained using the relation

$$\sigma = \frac{k}{\bar{v}_{\text{rel}}} \ , \qquad (3)$$

where $\bar{v}_{\text{rel}}$ is the average relative collision velocity.

The measured cross sections are shown in Fig. 1 together with the earlier results of Gallagher and Cooke[4] who suggest that, at low $n$, collisional depopulation proceeds via an interaction, of the $Na^+$ core with the target atom, which induces a transient dipole on the xenon atom that causes transitions to sodium states of different $n$. However, for the present range of $n$, the cross sections are very much larger, thereby suggesting that state-changing results from an interaction between the Rydberg electron and the target atom. In this event the amount of energy that can be transferred to or from a Rydberg atom by a collision with a xenon atom can be determined by considering the kinematics of an elas-

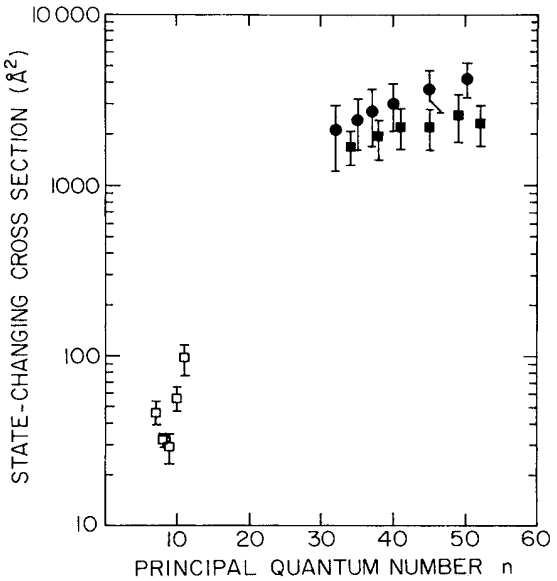


FIG. 1. Cross sections for state changing in Na**-Xe collisions. Present data, Na($np$) ●, Na($ns$) ■; Gallagher and Cooke (Ref. 4), Na($ns$) ⊟.

 

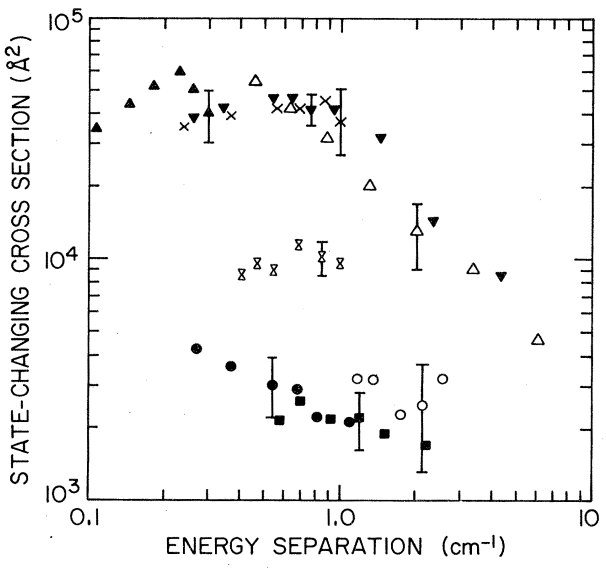


FIG. 2. State-changing cross sections as a function of the energy separation between the state of interest and the nearest manifold. Present data, Na($np$) ●, Na($ns$) ■; Kachru, Gallagher, Gounand, Safinya, and Sandner (Ref. 9), Na($nd$) ▼; Chapelet, Boulmer, Gauthier, and Delpech (Ref. 10), Na($nd$) ▲; Higgs, Smith, Dunning, and Stebbings (Ref. 6), Xe($nf$) ⧖; Hugon, Gounand, Fournier, and Berlande (Ref. 11), Rb($nf$) △; Hugon, Sayer, Fournier, and Gounand (Ref. 12), Rb($nd$) ⊕, Rb($ns$) ⧖.

tic collision between the Rydberg electron and the target atom. Such considerations show that small energy transfers are favored[1,2,8] and that, in consequence, transitions to the neighboring $n-1$ manifold should predominate, as is observed.

In Fig. 2 are shown the available data for state changing in collisions between various Rydberg atoms and xenon plotted as a function of the energy separation between each particular Rydberg state and the nearest manifold.[6,9–12] Of interest is the fact that these data fall into three distinct groups. The uppermost group pertains to quasihydrogenic $d$ and $f$ states for which state-changing collisions leave $n$ unchanged.[9–11] The lowest group comprises data for Na($ns$), Na($np$), and Rb($nd$) states which are not quasihydrogenic and which mix to the adjacent $n-1$ manifold. The data for Rb($ns$) states, which mix to the $n-3$ manifold, form the third group. This grouping according to the change in $n$ that occurs during state changing is entirely consistent with theoretical conclusions[1–3] that the degree of spatial overlap between the initial- and final-state wave functions is an important factor in determining the size of state-changing cross sections.

## ACKNOWLEDGMENTS

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