in its intensity due to mixing not only with the weaker $^1S_0$ levels, but also with the $6sng\,^1G_4$ series, which is not optically connected to the lower $6s6p\,^1P_1^\circ$ level. At the higher n values observed, further mixing with additional even-parity series is evidenced by the appearance of satellite lines and a rapid drop in oscillator strength as n increases.

In addition to the case of the $6s18s\,^1S_0$ level discussed earlier, the utility of the diamagnetic shift in differentiating between Rydberg states and doubly excited states is demonstrated by noting that the $6s6p\,^1P_1^\circ$-$5s7d\,^1D_2$ line in Fig. 3 exhibits a shift that is appreciably less than that of nearby $^1D_2$ Rydberg levels.

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Deflection of High-Rydberg Atoms in Collisions at Thermal Energy

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Lithium atoms in high Rydberg states, excited in an atomic beam by pulsed electron impact, are passed through a target gas and detected following electric-field state selection. Cross sections obtained from time-of-flight spectra are virtually independent of the principal quantum number and in numerical agreement with calculations for deflection of the core ion alone. This deflection has also been observed directly. These results suggest a new method for very-low-energy ion-atom scattering experiments.

Recent interest in the collisional interactions of high Rydberg states has been directed toward l-changing processes[1-3] and detachment of the outer electron by ions[4] and by polar and electronegative molecules.[5,6] When ionization takes place in collisions with molecules, the outer electron appears to behave as a free particle, independent of the core ion to which it is weakly bound. When Rydberg atoms are deflected by neutral atoms or molecules, it is conversely possible that the core ion is scattered as an electron-independent system.

Ion-atom and ion-molecule interactions have traditionally been studied by ion-beam scattering above about 1 eV and by drift-tube techniques at lower energies. The important thermal energy region has not been fully explored,[7] as slow ion beams have proved difficult to produce and maintain, while drift experiments measure ion transport properties rather than differential cross sections. This work considers the possibility that free-ion scattering can be observed at thermal energies in the deflection of high-Rydberg atoms by neutral gaseous targets. In the experiments to be described, a lithium atomic beam containing high Rydberg states (Li*) is scattered by a gas at low density.

The experimental geometry has been discussed previously.[8] Lithium atoms from a 500°C oven are excited by a transverse beam of electrons from a heated cathode. The electron accelerating potential is pulsed with a repetition rate of 850 Hz, a pulse width of 10 μsec, and an amplitude of 10 V. A 1.5-kG magnetic field, parallel to the electron beam, is applied to the excitation region to remove ions and electrons from the atomic beam and to focus the bombarding electrons.

Near the end of its 35-cm flight path, the Li beam passes through a pair of parallel electric field plates and a grounded aperture of 0.5-cm radius before entering an excited-atom detector.

The beam is collimated so that its edges approximately coincide with the detector aperture. Within the detector the Rydberg states are field ionized, and the stripped electrons are counted by a Channeltron electron multiplier. The resulting pulses are amplified, shaped, and then catalogued according to excited-atom flight time by a multichannel scaler. Dwell time per channel is 5.5 μsec; scan reset timing is synchronized with the excitation pulses. Even with an electron-source duty cycle of less than 1%, Li* counting rates in excess of 10^3/sec have been obtained. Target gas pressures up to 4×10^{-4} Torr are measured by a Bayard-Alpert ionization gauge calibrated by a Baratron capacitance manometer.

Time-of-flight spectra are obtained at various values of the electric field and target gas pressure. The detected states lie in a roughly defined interval of principal quantum numbers n. The maximum value $n_{max}=(E_0/E)^{1/4}$ is due to electric-field ionization[9] at the applied field E, where the constant E_0 is approximately 6×10^8 V/cm. A lower limit for the range of n values is determined by the initial population distribution and by radiative decay along the flight path. At a typical beam velocity of 2×10^5 cm/sec, the minimum lifetime of a Rydberg state which can survive without decay in flight is of order 10^{-4} sec. From measurements of Li* transit-time distributions[8] it is apparent that states of high orbital angular momentum are present in the beam transit region and that for $E>100$ V/cm the distribution of detected states receives a dominant contribution from principal quantum numbers just below n_{max}.

The 10-V cathode pulse amplitude provides almost twice the excitation energy required for Li* production, but is well below the ionization potential of the target gases used. Contributions to the signal from excitation of the target gas were not observed, and, in fact, no Rydberg atoms were detected with the oven unheated but with target gas present. Throughout the experiment, the Channeltron pulse-height distribution and the electron souce current (of order 0.3 mA during the pulse) were continuously monitored and found to be negligibly affected by the target gases, even at the highest experimental pressures.

The time-of-flight spectrum recorded at a base pressure of 2×10^{-6} Torr is given in the upper curve of Fig. 1, together with results obtained for a neon target at several partial pressures p. An applied electric field $E=200$ V/cm was maintained during these measurements. Introduction of the target gas causes the slower atoms to be preferentially scattered from the beam. Similar time-of-flight spectra were recorded for each of five target gases (He, Ne, Ar, H_2, and N_2) at electric fields $E=0$, 200, and 1000 V/cm. The corresponding average values of the principal quantum number in the detected beam are approximately $n=60$, 35, and 25.

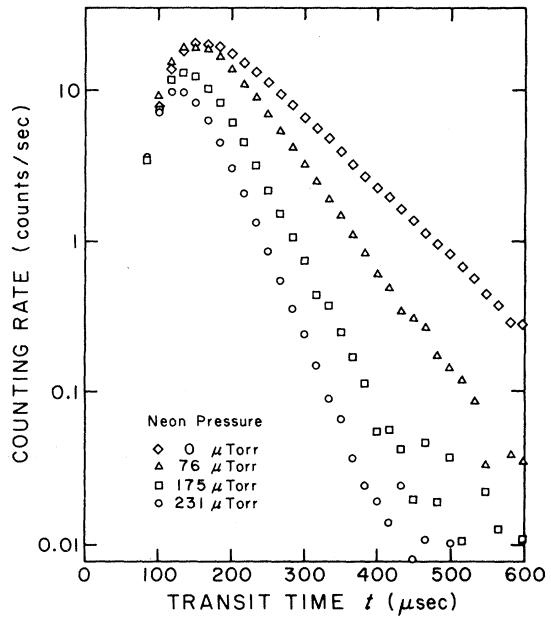

FIG. 1. Time-of-flight spectra for Li* atoms in a thermal beam, with neon as the target gas and an applied electric field $E=200$ V/cm. For display purposes each datum point represents a sum over three adjacent scaler channels.

For each target gas species and each electric field, a channel-by-channel ratio $R(t)$ was computed, in which the counting rate with the gas present was divided by the rate recorded in the same scaler channel without the target gas. The neon ratios are displayed in Fig. 2, which reveals nearly straight lines at large flight times t, on a semilog plot. The magnitudes of the slopes of these lines, denoted by γ, are plotted in Fig. 3 as a function of the target gas density n_0. The plotted points are consistent with a constant slope γ/n_0 which does not depend on E. Comparable results were obtained for the remaining target gases, for which the variation of slope with E appears to be random, without confirming the slight monotonic trend seen for neon in Fig. 3. The values of γ/n_0 are given in Table I.

If σ denotes a cross section for the removal of

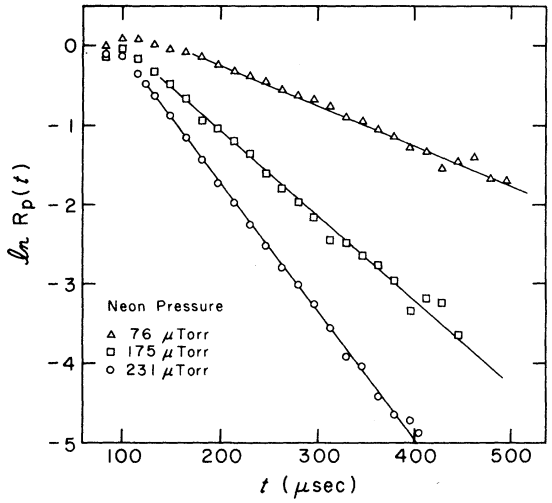

FIG. 2. The variation of $\ln R_p(t)$ with transit time t, for the data shown in Fig. 1.

Rydberg atoms (having velocity v) from the beam, then an exponential loss $R_p(t) = \exp(-n_0 \sigma v t)$ is expected in the absence of radiative decay and collision-induced changes of state. The latter two processes can account for the overall displacement of the experimental plots of $R_p(t)$ (Fig. 2) as well as the deviation from straight-line behavior at low channel numbers. These effects are due to the relatively rapid radiative decay of the initially excited low-l states and the much slower radiative decay of the higher-l states which are populated as a result of l-changing collisions. It is then possible to make the identification $\gamma/n_0 = \sigma v$. A velocity dependence

$$\sigma \propto 1/v \qquad (1)$$

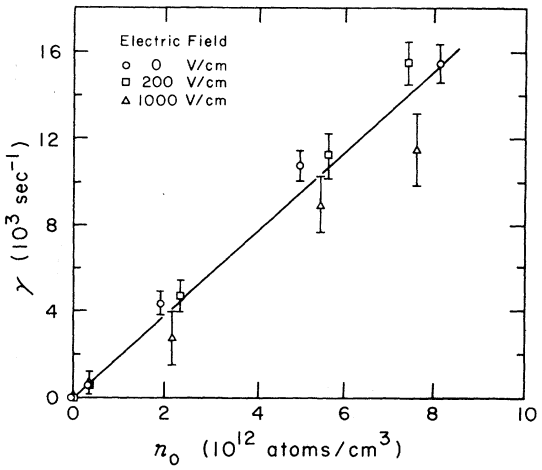

FIG. 3. Experimental values of γ, plotted vs neon density n_0. The slope γ/n_0 is entered as σv in Table I.

TABLE I. Experimental and calculated values of σv for the five target gases.

Target gas	α/a_0^3	Experimental $\sigma v = \gamma/n_0$ (10^{-9} cm^3/sec)	Calculated σv (Eq. 4) (10^{-9} cm^3/sec)
He	1.38	1.97 ± 0.40	1.75
Ne	2.67	1.88 ± 0.45	2.42
Ar	11.07	2.37 ± 0.65	4.94
H_2	5.45	4.00 ± 0.65	3.47
N_2	11.88	2.12 ± 0.60	5.12

is implied by the observation of an essentially time-independent γ in Fig. 2 and in the analysis for the other target gases.

The lack of E dependence suggests that σ may describe a localized scattering process for the core ion alone. Since alkali absorption transitions to high-n states have been observed even with large numbers of rare-gas atoms present within the Rydberg orbit radius,[10] it is reasonable to suppose that a target atom can penetrate the outer electron cloud and transfer momentum to the core ion. For a direct test of this deflection hypothesis, a movable collimating slit, 3 mm in diameter, was inserted into the atomic beam path, 7 cm from the source. The Li* beam profile was scanned and, as shown in Fig. 4, a pronounced spreading was observed as the target gas pressure was increased. Within experimental error, the beam area in the detector plane is inversely proportional to the on-axis counting rate. The spreading is thus sufficient to account for the central beam attenuation, without deple-

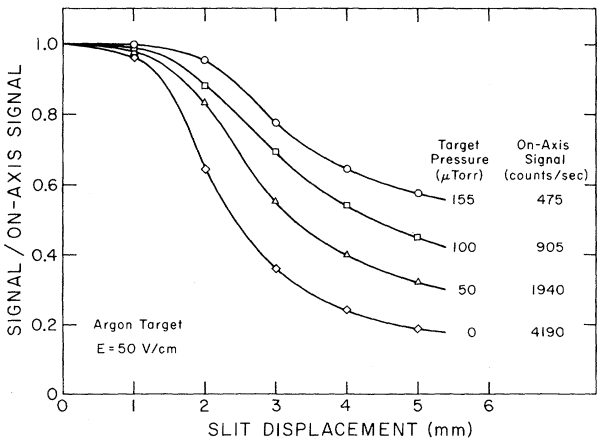

FIG. 4. Direct observation of Li* deflection by means of a movable collimating slit.

tion of the Li* population by electron-dominated processes such as collisional deexcitation or ionization.

The elastic scattering of a slow ion by an atom having polarizability α is described by an attractive polarization potential having the asymptotic form $V(r) = -\alpha e^2/2r^4$. For this potential a classical calculation of the differential cross section for small scattering angles θ (in the laboratory frame) yields

$$d\sigma/d\Omega \cong v^{-1}(3\pi\alpha e^2/64m\theta^5)^{1/2} \quad (2)$$

after integration over the Maxwell-Boltzmann velocity distribution for the target. The transformation of the scattering angle from the center-of-mass reference frame accounts for the presence of the lithium mass m, rather than the reduced mass, in Eq. (2). Integration over scattering angles and interaction locations along the beam leads to an estimate of the observed scattering cross section:

$$\sigma = \frac{R}{L}\int_{R/L}^{\varphi'} \frac{d\varphi}{\varphi^2}\int_{\varphi}^{\theta'} \frac{d\sigma}{d\Omega} 2\pi \sin\theta \, d\theta . \quad (3)$$

Here L and R represent the beam length and the radius of the detector aperture, respectively, and φ denotes the half-angle intercepted by the detector at an arbitrary point along the beam trajectory. Angles φ' and θ' are large and do not appear in the result,

$$\sigma \cong v^{-1}(\pi^3\alpha e^2 L/3mR)^{1/2}, \quad (4)$$

which is consistent with Eq. (1). Table I includes numerical values of σv calculated from this relation for the experimental dimensions $L = 35$ cm and $R = 0.5$ cm. Typical cross sections are of order 10^{-14} cm^2.

The experimental and calculated values of σv are in excellent agreement for He, Ne, and H_2, and are within a factor of 2.5 for the most massive targets, Ar and N_2. This agreement gives further support to the free-ion view of the scattering process. Equation (4) does not take into account quantum scattering effects, which become important at the smallest of the observed deflections. Short-range corrections to $V(r)$, which affect large-angle scattering, are likewise not included in this calculation.

There are interesting prospects for further experimental work, including the measurement of differential scattering cross sections and the study of ion participation in chemical reactions. High-Rydberg atoms may thus play a useful role in velocity-resolved ion-atom and ion-molecule scattering experiments at thermal energy. The presence of a Rydberg-state valence electron provides a charge-neutralizing sheath around the ion, rendering its trajectory virtually unaffected by nonionizing external fields.

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