ments of β_4 have involved measurements of transition strengths, e.g., $B(E4, 0^+ \rightarrow 4^+)$, and have therefore not established that there exist static $E4$ deformations. The results of our spectral calculations provide evidence that indeed the existence of such $E4$ transitions results from static $L=4$ deformations rather than, e.g., $L=4$ vibrations.

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Absolute Measurements of Collisional Ionization of Xenon Atoms in Well-Defined High Rydberg States*

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Measurements of the absolute cross sections for Xe^+ production in collisions between xenon atoms in a single well-defined high Rydberg state $|nf\rangle$ and SF_6 are reported and are $\sim 10^{-11}$ cm^2 for $25 \leq n \leq 40$. The data lend support to the hypothesis that these collisions are dominated by the electron-SF_6 interaction, with the Xe^+ ion core playing a minor role.

Atoms in high Rydberg states with principal quantum number $n \geq 25$ have ionization potentials ≤ 20 meV. As a result sufficient energy is available in thermal encounters for collisional ionization of such atoms to occur. Earlier studies of collisional ionization[1-3] using beams of high Rydberg atoms with an unknown mixture of n in the range 20 to 100 yielded cross sections of the order of 10^{-12} cm^2 when the target was a polar molecule, or had a large thermal electron-capture cross section. Theoretical work[4] shows that for such targets, cross sections of this order of magnitude are to be expected.

Recent experiments in this laboratory[5] and elsewhere[6-8] have shown that atoms in selected high Rydberg states can be produced by optical excitation and detected absolutely using field ionization. In the present experiment these techniques have been used to measure absolute cross sections for the ionization, in collision with SF_6, of xenon atoms in well-defined high Rydberg states, represented in jl notation as $nf[\frac{3}{2}]_1$ with

n in the range $25 \leq n \leq 40$. The collision may be represented by

$$Xe(nf) + SF_6 \rightarrow Xe^+(^2P_{3/2}) + (SF_6 + e). \quad (1)$$

The apparatus has been described in detail elsewhere[5,9,10] and only the essential features are described here. A beam of xenon atoms, a small fraction of which were excited to the metastable $^3P_{0,2}$ states by electron impact was directed into a region containing the SF_6 target gas at a pressure of $\sim(1-2)\times10^{-6}$ Torr, a factor of about 30 above the background gas pressure. A fraction of the $Xe(^3P_0)$ atoms in the beam was then excited, in zero electric field, to a selected high Rydberg state using a pulsed tunable dye laser. The atoms so formed have radiative lifetimes, τ, of several microseconds[5] and so their subsequent loss resulted from a combination of spontaneous decay and collisional ionization. As a consequence the expression relating the rate constant to the experimental parameters which is normally used for thin-target conditions[11] is not applicable and instead the collisional-ionization rate constant k is determined from the expression

$$S(t) = N(0)n[1 - \exp(-t/\tau)]\tau k, \quad (2)$$

where n is the number density of the SF_6 and is determined using an ion gauge calibrated against an MKS Baratron pressure gauge.[12] $S(t)$ is the number of Xe^+ ions formed in ionizing collisions during a period t (≈ 4 μsec) following the laser pulse and is determined by sweeping them to a Johnston particle multiplier with an electric field (~ 40 V/cm) applied immediately after this interval. $N(0)$, the number of high Rydberg atoms produced during the laser pulse, is obtained from

$$N(0) = N(t')e^{t'/\tau}, \quad (3)$$

where $N(t')$ is the number of high Rydberg atoms present at a time t' (≈ 1.0 μsec) after the laser pulse and was determined by use of the field-ionization technique.[5] It is evident from the nature of these measurements that following any laser pulse it is only possible to measure either $S(t)$ or $N(t')$. Because the determination of both $S(t)$ and $N(t')$ requires the detection of Xe^+ ions, it is clear from Eq. (2) that the absolute detection efficiency of the multiplier which was used for both measurements need not be known. A gating technique was used for both measurements to discriminate against ions formed through chemi-ionization of the residual gas by xenon metastables in the beam. Penning ionization of the SF_6 by both xenon metastable and high Rydberg atoms is not energetically possible and a time-of-flight analysis showed that associative ionization processes involving SF_6 made an immeasurably small contribution to the ion signal. Chupka[8] has also shown that chemi-ionization in collisions of highly excited rare-gas atoms with SF_6 is several orders of magnitude less probable than collisional ionization. Thus the signal $S(t)$ is due to xenon ions produced through process (1).

Of concern was the possibility that during the time interval t the high Rydberg atoms might suffer collisions which would change their quantum state[7] and thus destroy the purity of the beam. A variety of tests involving field ionization and the dependence of signal on pressure and t were therefore carried out but gave no evidence that collisions occurring in this time interval were important in this respect.

The measured rate constants for process (1) are shown in Table I together with cross sections, σ, obtained from the approximate relation $\sigma = k(\bar{v}_{rel})^{-1}$ where $\bar{v}_{rel}$ is the average velocity of the xenon high Rydberg atoms relative to the target molecules.[13] The total uncertainties of these quantities are given although the uncertainty in relative values is considerably less ($\sim 12\%$). These cross sections are among the largest yet observed for thermal-energy neutral-neutral collisions and it could be argued that this is simply a consequence of the large physical size of the high Rydberg atoms. However as n increases the atoms become progressively larger (radius $\propto n^2$) and the outer electron becomes less strongly bound (binding energy $\propto n^{-2}$). Both effects would be expected to cause the collisional ionization cross section to increase with n, and the fact that σ is observed to be insensitive to the value of n suggests that this simple picture is inappropriate.

An unexpected, and so far unexplained, effect is that the rate constants are influenced by the application at the interaction region of an electric field subsequent to the exciting laser pulse. Figure 1 shows the observed variation of k with field strength for atoms excited to states $|25f\rangle$ and $|28f\rangle$. Earlier measurements[5] showed that the radiative lifetimes of these atoms are appreciably lengthened in electric fields of this magnitude and account of this effect is taken in the evaluation of the rate constants from Eq. (2). In an effort to shed further light on this phenomenon preliminary studies have been made with target species other than SF_6 and their collisional-

TABLE I. Rate constants and cross sections for collisional ionization in collisions with SF_6.

High Rydberg atom	Rate constant, k (10^{-7} cm^3 sec^{-1})	Cross section, σ (10^{-11} cm^2)
Xe $25f$	3.7 ± 0.7	1.1 ± 0.2
Xe $26f$	4.0 ± 0.8	1.2 ± 0.2
Xe $28f$	3.8 ± 0.8	1.2 ± 0.2
Xe $31f$	3.6 ± 0.7	1.1 ± 0.2
Xe $33f$	4.2 ± 0.7	1.1 ± 0.2
Xe (35 ± 1)[a]	3.8 ± 0.8	1.3 ± 0.3
Xe (38 ± 1)[a]	4.0 ± 0.8	1.2 ± 0.2
Xe (40 ± 2)[a]		1.2 ± 0.2
Ar[b] $20 \leq n \leq 100$		0.15 ± 0.01[c]
		0.17 ± 0.01[d]

[a] For $n > 34$ the angular momentum state is not determined Ref. 5 but is probably f.

[b] Reference 1, measurement made in a field of 100 V/cm.

[c] From observations of Ar^+ production.

[d] From observations of SF_6^- production.

ionization cross sections were found to exhibit small or zero E-field dependences. It is nonetheless evident that this effect must be taken into account in collisional-ionization studies, and in this connection it should be noted that the earlier measurements of Hotop and Niehaus,[1] presented in Table I, for argon high Rydberg atoms incident on SF_6 were made in an electric field of 100 V/cm.

Conventionally the collision represented by Eq. (1) is viewed as occurring between two atomic systems. However Matsuzawa[4] has proposed a model of this collision in which, because of the large separation of the excited electron from its accompanying ion core, the effect of the latter on the collision may be essentially ignored. In this event the collision may be viewed simply as occurring between the SF_6 molecule and the highly excited electron whose energy is that of its orbital motion. Developing this model further Matsuzawa shows that the rate constant for electron transfer from a high Rydberg atom A^* to a molecule B at thermal energies,

$$A^* + B \rightarrow A^+ + B^-, \tag{4}$$

should be equal to the rate constant for attachment of free electrons[14] of the same energy to B,

$$e + B \rightarrow B^-. \tag{5}$$

In the present work the fate of the detached electron is not determined but similar studies of collisions of high Rydberg atoms with SF_6 by Hotop and Niehaus[1] and by Chupka[8] show that when collisional ionization occurs the Rydberg electron is captured by the SF_6 molecule. Assuming similar behavior in the present work the rate constants for collisional ionization of high Rydberg xenon atoms should be equal to the rate constants for the attachment of free thermal electrons, of equivalent energy, to SF_6.

To test this hypothesis the present results are compared in Fig. 2 with data[15,16] for attachment of free electrons to SF_6. The agreement is remarkably good although no detailed comparison

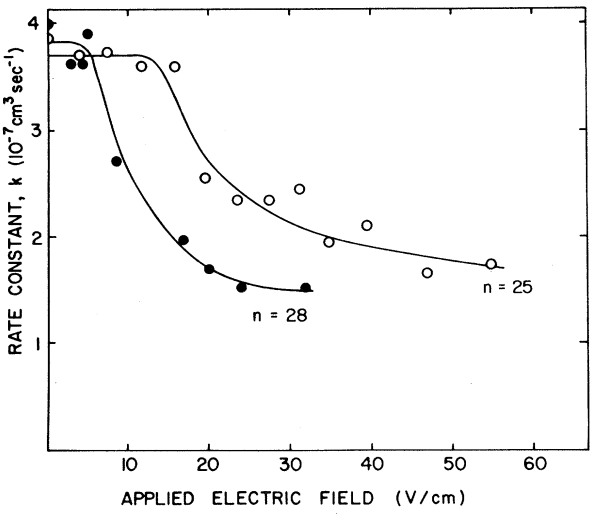

FIG. 1. Rate constants for collisional ionization of Xe atoms excited to $|25f\rangle$ and $|28f\rangle$ states in collisions with SF_6 as a function of the electric field applied at the interaction region subsequent to each laser pulse.

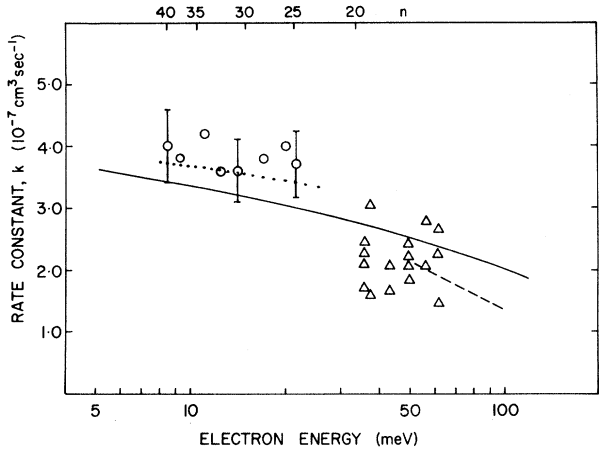

FIG. 2. Rate constants for electron attachment to SF_6 as a function of average electron kinetic energy. ○, present data: electrons bound in xenon (nf) high Rydberg atoms. ---, Christophorou *et al.*, 1971: electron-swarm experiment. △, Fehsenfeld, 1970: flowing-afterglow experiment. —, theory: a Maxwellian velocity distribution. •••, theory: velocity distributions appropriate to Xe(nf) Rydberg electrons.

is possible since there is no overlap in energy. To further this comparison Fig. 2 includes rate constants for free-electron attachment to SF_6 which are derived from

$$K = \int_0^\infty v\sigma_c(v) f(v) dv, \qquad (6)$$

where $\sigma_c(v)$ are theoretical cross sections[17] for process (5). If a Maxwellian velocity distribution is used for $f(v)$ the agreement with all the experimental results is seen to be reasonably good (although it must be noted that the electron energy distribution in a high Rydberg orbit of low angular momentum is not Maxwellian). However Percival and Richards[18] have given expressions which may be used to calculate the electron energy distributions for classical hydrogenic orbits of angular momentum $l = 3$ and when these distributions are used in Eq. (6), the resulting theoretical rate constants are seen to be in excellent accord with the present data.

The present measurements therefore lend support to Matsuzawa's model of the collisional-ionization process for target molecules which have large thermal-electron capture cross sections. It should be noted that in this model the physical size of the Rydberg atoms is of no significance and no rapid increase in σ with n, as discussed earlier, is to be expected. A small increase of σ with n is however predicted (Fig. 2) and is consistent with the experimental data.

In conclusion the results indicate that experiments with high Rydberg atoms may, in certain circumstances, be used to provide information on electron scattering in the experimentally difficult region below 20 meV. It is interesting to speculate also that certain target species may interact more strongly with the ionic core of a high Rydberg atom than with the outer electron. In this event such collisions could provide a useful new technique for the study of thermal-energy ion-molecule reactions.

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$$\frac{d[B^-]}{dt}=k[A^*][B],$$

where k is the rate constant and $[A^*]$ and $[B]$ are the concentrations of A^* and B, respectively. However if the ion core of the A^* atom plays no part in the interaction we may write this as

$$\frac{d[B^-]}{dt}=k[e][B],$$

since $[A^*]=[e]$. But this is just the rate of electron attachment to molecule B [process (5) in text].

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Single and Double Charge Transfer in C^{4+}-He Collisions*

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Cross sections for single and double charge transfer from C^{4+} incident on He have been measured and calculated for relative velocities between (0.2 and 1.2) $\times 10^8$ cm/sec. The double-charge-transfer cross section rises with decreasing energy and is predicted to be nearly two orders of magnitude larger than that for single charge transfer at the lowest velocity. The single- and double-charge-transfer cross sections are experimentally found to be equal at a velocity of 6.5×10^7 cm/sec with a value of 2.7×10^{-16} cm^2.

Multiply charged ions of common atoms are of considerable interest in high-temperature and nonequilibrium plasmas. Because of large cross sections, charge exchange between these ions and other atomic constituents in the plasmas plays a significant role in important plasma properties such as charge-state distribution, resistivity, and energy transfer. In the interstellar medium, charge exchange is important in determining which ion states are expected.[1] In tokamak plasmas charge exchange of multiply charged ions is important for the properties mentioned above; in addition, for injected neutral beams, charge exchange with multicharged impurities may be instrumental in stopping the injected beam at the outer edge of the plasma, thus destroying the needed plasma heating.[2] An additional area where such charge-transfer cross sections are important is in the search for x-ray lasers, where charge exchange into an excited state of a multicharged ion could provide a mechanism for pumping the required excited-state population.[3]

Because of these several important areas of application, it is necessary to develop better quantitative knowledge of the collision processes involving multiply charged ions. In the case of charge exchange at relatively slow velocities, little experimental work exists and dependable theoretical predictions are also scarce; but both experiments[4] and theory are actively being pursued. In the course of recent investigations, some of the qualitative ideas held in general use for estimating effects of charge exchange with multicharged ions have proven to be false in many cases.

Usually, it is assumed that for slow collisions