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LETTER TO THE EDITOR

Ionisation of fast Rydberg ions in collision with target atoms

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Abstract. Rydberg ions with orbital dimensions up to approximately 1000 a_0 are found to traverse thin gas targets fairly undisturbed, indicating relatively small ionisation cross sections σ_i. Theoretical estimates which take into account screening of target nuclei reveal that $\sigma_i(n)$ becomes asymptotically independent of principal quantum number n.

In recent years a number of reports have dealt with Rydberg ions which travel through gaseous targets and residual gas in beam lines prior to being observed by field ionisation or radiative decays (Braithwaite *et al* 1975, Kim and Meyer 1980, Betz *et al* 1980, 1983). It is well understood that the long lifetime of Rydberg states, $\tau_{nl} \propto n^3 l^2$, where n and l denote principal and angular quantum number, respectively, allows detection long after creation of the state, but no explanation was given for the fact that these ions could be observed and were not destroyed in collisions with atoms along the flight path. For obvious reasons, a steep rise of electron-loss cross section, $\sigma_i(n) \propto n^2$, is expected only for ionisation by *charged* particles. We estimate ionisation by *neutral* atoms and find that $\sigma_i(n)$ deviates significantly from a n^2 dependence even for the lowest n values and becomes asymptotically independent of n. The latter result is in agreement with expectations from Butler and May (1965) and estimates by Matsuzawa (1980).

125 MeV sulphur ions from the Munich Tandem van de Graaff accelerator were prestripped to obtain bare nuclei (charge 16^+) and directed through a gas cell of length 3 cm containing target gases (N_2, CH_4) at pressures up to approximately 1 Torr. Two cryogenic pumping systems aided by two turbomolecular pumps served to achieve high vacuum (about 10^{-6} Torr) behind the gas target. At a distance of 10 cm behind the centre of the gas target a Si(Li) detector observed K x-rays due to decays of Rydberg states formed by electron capture in the gas target. Energy resolution was sufficient to resolve hydrogen-like transitions. Separate experiments with other lengths of the gas cell and variation of target–detector distance showed that the observed x-ray decays contain negligible contributions due to states formed by collisions along the flight path between gas target and the end of the detection region. Beam intensity was monitored by means of a Faraday cup. Further experimental details and discussions of the technique to observe Rydberg states via radiative decay cascades are summarised in a forthcoming paper (Röschenthaler *et al* 1983).

Figure 1 presents the absolute intensity of Ly-α (2p$\rightarrow$1s) and 2E1 (2s$\rightarrow$1s) transitions measured 10 cm behind the gas target, as a function of the target thickness x. The Ly-α intensity signifies cascaded decays of hydrogen-like Rydberg states with initial quantum numbers as high as $n \simeq 100$, which corresponds to orbital dimensions of approximately 1000 a_0, whereas the 2E1 intensity measures the population of the metastable 2s state of hydrogen-like sulphur ions. Both curves increase linearly for small target thicknesses indicating that single-collision conditions prevail. Saturation effects become evident near $x_\alpha \simeq 3\times10^{16}$ and, with some uncertainty, near $x_{2s} \simeq 6\times10^{16}$ atoms/cm^2 for the Ly-α and 2E1 curves, respectively.

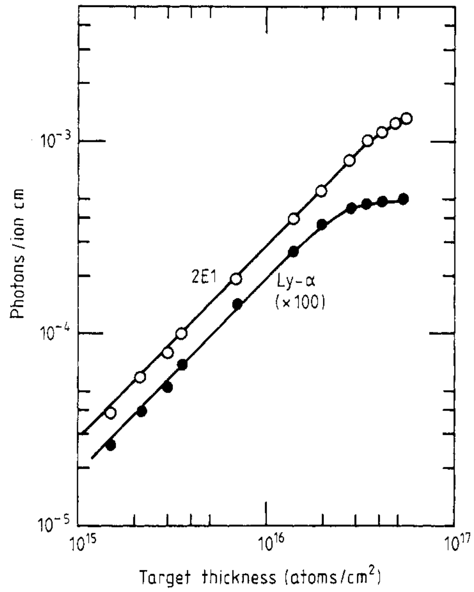

Figure 1. Absolute Ly-α and 2E1(2s$\rightarrow$1s) intensities of sulphur ions, observed 10 cm behind a N_2 gas target, as a function of target thickness. The Ly-α intensity shown signifies decay of long-lived Rydberg states.

General consideration of charge exchange for varying target thickness reveals that a linear relationship as shown in figure 1 must break down when a target thickness $x \simeq \sigma_t^{-1}$ is reached, where σ_t is the *sum* of cross sections for production and destruction of the considered state. We conclude, therefore, that the ionisation cross section for any Rydberg state does not exceed approximately 3×10^{-17} cm^2 in the present collision system.

In order to calculate electron loss from hydrogen-like ions in collisions with neutral atoms we use the free-electron model (Matsuzawa 1980) in which the total cross section is given in terms of the ionisation matrix element of the projectile and the amplitude $f_e^T(\boldsymbol{q})$ for the scattering of the projectile electron by the target atom (in atomic units)

$$\sigma_i = v^{-1} \int \mathrm{d}\boldsymbol{k}_f \int \mathrm{d}\boldsymbol{q}\, |\langle\psi_f^P|\exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r})|\psi_i^P\rangle|^2 |f_e^T(\boldsymbol{q})|^2 \delta(E_f - E_i + \boldsymbol{q}\cdot\boldsymbol{v}) \qquad (1)$$

where ψ_i^P and ψ_f^P are the intial and final electronic states of the projectile with energies

E_i and $E_f = k_f^2/2$, respectively, and the projectile moving with velocity v is used as reference frame. When we neglect target resonances and excitation f_e^T is obtained from the first Born approximation, i.e. from the Fourier transform of the screened target field, $V(r) = -(Z_T/r)\exp(-r/d)$, with screening length d and nuclear charge Z_T of the target,

$$f_e^T(q) = -(2\pi)^{1/2} V_T(q) \simeq 2Z_T/(q^2 + d^{-2}). \tag{2}$$

With this, σ_i reduces to the first-order Born approximation for ionisation. When we average over quantum numbers l and m a closed expression is obtained for the matrix element which we have used to calculate cross sections for $n < 30$ without screening (figure 2, curve 1), and utilising the Thomas–Fermi approximation $d^{-1} = 1.13\, Z_T^{1/3}$ (figure 2, curve 3). For the latter case $\sigma_i(n)$ is seen to approach an n-independent value for large n.

Almost identical results for $\sigma_i(n)$ can be derived more directly from the plane-wave Born approximation as formulated, for example, by Madison and Merzbacher (1975) applied to hydrogenic 1s-type wavefunctions, where the nuclear charge of the projectile, Z_p, is replaced by Z_p/n. Screening can be implemented by replacing dq/q by

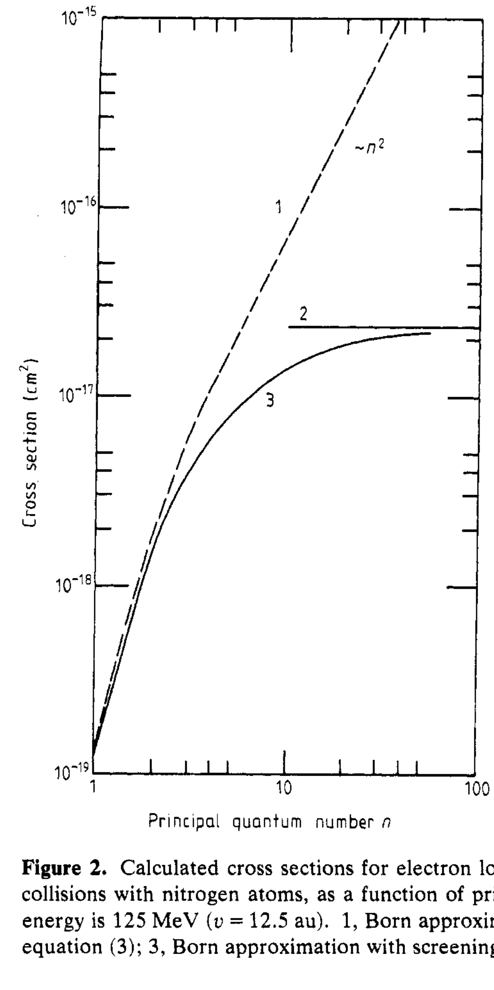

Figure 2. Calculated cross sections for electron loss from hydrogen-like sulphur ions in collisions with nitrogen atoms, as a function of principal quantum number n. Projectile energy is 125 MeV ($v = 12.5$ au). 1, Born approximation without screening ($Z_T^{eff} = 7$); 2, equation (3); 3, Born approximation with screening (see text).

$dq\, q^3/(q^2+d^{-2})^2$ in the integration over momentum transfer (Drepper and Briggs 1976).

An analytic expression for the asymptotic cross section $\sigma_i(\infty)$ can be derived from the above when the final electron wavefunction is taken as a plane wave, along with further simplifications such as the peaking approximation. A simpler and more direct method to obtain $\sigma_i(\infty)$ is to treat the collision as elastic (Rutherford) scattering of a free electron on the screened potential $V(r)$, whereby one must take into account that momentum transfer is restricted to $q > q_{\min} = \Delta E/v = Z_p^2/(2n^2 v)$ (Bell 1982). In both cases we get the simple relation

$$\sigma_i(n) = \frac{4\pi Z_T^2}{v^2 d^{-2} + Z_p^4 (4n^4)^{-1}} \xrightarrow[n\to\infty]{} \frac{4\pi Z_T^2 d^2}{v^2}. \tag{3}$$

This result is displayed in figure 2 (curve 2) and gives the same asymptotic cross section as the more rigorous procedure (curve 3). We note, however, that equation (3) is not intended for small values of n and becomes invalid for vanishing screening. Interestingly, equation (3) yields a characteristic saturation value n_0, defined by $\sigma_i(\infty) = 2\sigma_i(n_0)$, which can also be obtained by equating d with the adiabatic distance $b_{ad} = q_{\min}^{-1}$,

$$n_0 = Z_p (d/2v)^{1/2}.$$

For the present collision system we estimate $d = 0.46$ and get $\sigma_i(\infty) = 2.4 \times 10^{-17}\ \mathrm{cm}^2$ although it must be kept in mind that a molecular target (N_2) may have to be described by a somewhat larger screening radius which implies larger values σ_i.

Destruction of a hydrogen-like Rydberg state is achieved not only by electron loss, but also by capture of a further electron into any projectile state. Appropriate total cross sections can be obtained, for example, from the Eikonal approximation (Eichler 1981) which yields $\sigma_c^T = \Sigma_n \sigma_c(\to n) \simeq 10^{-17}\ \mathrm{cm}^2$. Production of Rydberg states from bare nuclei also proceeds via electron capture, but since only a single final state is relevant, this cross section must be small compared with σ_c^T. Altogether, the resulting sum of cross sections for production and destruction of the observed Rydberg states becomes $\sigma_i(\infty) + \sigma_c^T \simeq 3.4 \times 10^{-17}\ \mathrm{cm}^2$, and is in reasonable agreement with our experimental estimate x_α^{-1} derived from the Ly-α data.

As regards the 2E1 data we apply a similar interpretation; destruction of the 2s configuration is due to electron loss, $\sigma_i(2) = 1.7 \times 10^{-18}\ \mathrm{cm}^2$ (figure 2, curve 3), and capture of any other electron, $\sigma_c^T \simeq 10^{-17}\ \mathrm{cm}^2$, whereas production results from electron capture into the 2s and several higher states cascading to the 2s state and is small compared with σ_c^T. The sum $\sigma_i(2) + \sigma_c^T \simeq 1.2 \times 10^{-17}\ \mathrm{cm}^2$ compares well with the estimate from the 2E1 data, $x_{2s}^{-1} \simeq 1.7 \times 10^{-17}\ \mathrm{cm}^2$.

Effects which remain to be clarified concern the action of the projectile nucleus on the target atom and projectile ionisation due to electron–electron interaction. Our data seem to suggest that these effects do not give dominant contributions to Rydberg ionisation.

Finally, we would like to point out another consequence of asymptotically constant ionisation cross sections. The n distribution of Rydberg electrons produced, for example, by electron capture in ion–atom collisions ($\propto n^{-3}$) will *not* change when the ions undergo multiple collisions and attain charge equilibrium, in contrast to previous conjectures (Hopkins and Brentano 1976, Latimer 1982).

In conclusion, we note that calculation of projectile ionisation by neutral target atoms (including passage through solids) requires consideration of screening effects as discussed above even for moderate values of n.

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