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LETTER TO THE EDITOR

Electron capture from elliptic Rydberg states

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Abstract. Recent experiments on electron capture from elliptic Rydberg states by Ehrenreich *et al* have shown that the total electron capture cross section depends strongly on the eccentricity, e, of the elliptic state and the orbit direction of the electron. By carrying out close-coupling calculations for protons colliding with elliptic H ($n = 3, 4, 5$) atoms we show that the dependence of total capture cross section on e varies weakly with the principal quantum number n. We also present the dependence of the total capture cross section as function of the angle, φ, between the incoming ions and the classical plane of the elliptic orbital. Both the dependence on e and on φ are shown to be consistent with the criteria of velocity matching for the electron capture process.

In a recent paper Ehrenreich *et al* (1994) reported the measurement of total electron capture cross section in collisions between 2.5 keV Na ions and Li atoms in an elliptic state with principal quantum number $n = 25$. The elliptic state is created in crossed electric and magnetic fields (Day *et al* 1994), where the eccentricity, e, of the elliptic state is determined by the ratio of the magnetic field, B, to the electric field, E. The limiting cases $B = 0$ and $E = 0$ correspond respectively to the extreme Stark state, $e = 1$, and to the circular state, $e = 0$. The latter case has been studied experimentally by Hansen *et al* (1993) and theoretically by Wang and Olson (1994) and by Lundsgaard *et al* (1994). In this way the eccentricity and the orientation of the electron's orbit can be varied continuously by changing the field strengths, making a detailed study of the dynamical properties of the collision system possible even at the level of total capture cross section. In the experimental setup, the electric field is always perpendicular to the beam axis while the direction of the B-field with respect to the beam axis can be changed continuously (Hansen *et al* 1993), which introduces a second parameter to the system, the so-called angle of orientation (φ).

An elliptic state with energy E_n has its spatial electron distribution centred on an ellipse and mimics in this sense a classical Kepler orbital. Also the momentum distribution of the electron is similar to its classical counterpart, i.e. it peaks on a circle that is centred not at the nuclei, but at a point shifted away from the nuclei along the minor axis by a distance proportional to e. The elliptic state is characterized by the expectation values of the angular momentum, L, and the Runge–Lenz vector A (Gay *et al* 1989)

$$\langle L_z\rangle = \pm(n-1)\sqrt{1-e^2} \qquad \langle L_x\rangle = \langle L_y\rangle = 0$$
$$\langle A_x\rangle = (n-1)e \qquad \langle A_y\rangle = \langle A_z\rangle = 0. \tag{1}$$

The sign of $\langle L_z\rangle$ determines the orientation of the elliptic state.

In this letter we study the dependence of electron capture cross sections on the eccentricity and the orientation of the elliptic states in ion–atom collisions using the close-coupling method. Such a calculation can still not be carried out if the initial elliptic state is at $n = 25$. We will show results for calculations carried out for elliptic states where the principal quantum number n ranges from 3 to 5 and examine how the total electron capture cross sections depend on the eccentricity e and the orientation angle φ.

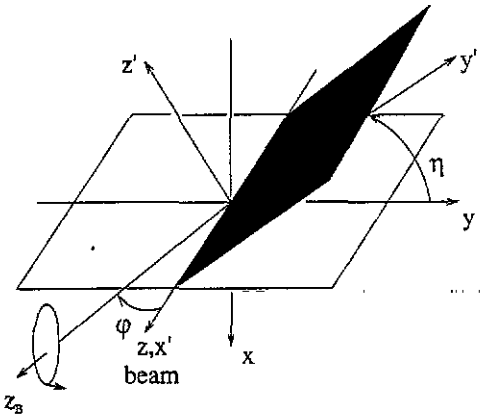

Figure 1. The geometry for describing the collisions. The ion beam is travelling in the direction of z (and x'). The xyz is a space-fixed frame of reference. The scattering plane is the $x'y'$ plane which makes an angle η with respect to the space-fixed xz plane. z_B is the direction in which the elliptic state is oriented; it makes an angle φ with respect to the beam.

We describe the collision in the so-called '*natural frame*' of reference in which the collision plane is defined as the $x'y'$ halfplane with y' positive and with the incident beam moving in the $+x'$ direction (see figure 1). Thus the quantization axis $+z'$ is perpendicular to the collision plane. The elliptic state, $|n, e\rangle$, created in the experiment is defined in the xyz frame of reference, where the electric field $\boldsymbol{E}$ is pointing in the $-x$ direction and the beam in the $+z$ direction. The magnetic field is always perpendicular to $\boldsymbol{E}$, but makes in general an angle φ with the beam axis. The $|n, e\rangle$ state created in the experiment can be obtained from the corresponding elliptic state, $|n, e\rangle'$, defined in the natural frame of reference, by two successive rotations,

$$|n, e\rangle = D(\eta + \pi, \tfrac{1}{2}\pi, \pi)\, D(\tfrac{1}{2}\pi, \varphi, -\tfrac{1}{2}\pi)\, |n, e\rangle'. \tag{2}$$

Here D is the rotation operator as defined in Edmonds (1960). The angle η determines the orientation of the scattering plane, while φ is the angle between $\boldsymbol{B}$ and the incoming beam, see figure 1.

Since the wavefunctions in all the close-coupling calculations are expressed in terms of spherical coordinates, it is convenient to expand $|n, e\rangle'$ in terms of the substates of the n-manifold (Gay *et al* 1989)

$$|n, e\rangle' = \sum_{lm} c_{nlm}(e) |nlm\rangle'$$

$$c_{nlm}(e) = (-1)^{(l+m)/2} \frac{2^{n-l-1}(n-1)!}{(\frac{1}{2}(l-m))!(\frac{1}{2}(l+m))!} \left(\frac{(l+m)!(l-m)!(2l+1)}{(n-l-1)!(n+l)!} \right)^{1/2} \times (\sin\tfrac{1}{2}\alpha)^{n-m-1} (\cos\tfrac{1}{2}\alpha)^{n+m-1} \qquad \sin\alpha = e. \tag{3}$$

Because the elliptic state has even reflection symmetry with respect to the collision plane, the summation in equation (3) above only extends over even substates of the n-manifold. The analytical dependence on e, φ, and η of the amplitude, a_{fi}, for the transition $|i\rangle \rightarrow |f\rangle$ with $|i\rangle = |n, e\rangle$ may now be obtained from equations (2) and (3),

$$a_{fi}(e, \varphi, b, \eta) = \sum_{lmqp} \mathrm{i}^m (-1)^p d^{(l)}_{pq}(\tfrac{1}{2}\pi)\, d^{(l)}_{qm}(\varphi)\, c_{nlm}(e) \mathrm{e}^{\mathrm{i}q(\eta+\pi)} a'_{f,lp}(b). \tag{4}$$

The amplitudes $a'_{f,lp}(b)$ refer to the transition $|nlp\rangle' \rightarrow |f\rangle$, where the dependence of the transition amplitude on the impact parameter, b, is indicated. By integrating the transition probability over all impact parameter planes and summing over all final states, we obtain the total capture cross section

$$\sigma_{\mathrm{cap}}(e, \varphi) = \sum_{lp,l'p'} \Big(\sum_{mm'} \lambda'_{lm,l'm'}(n; e) G(lp, l'p', m, m'; \varphi)\, g \Big) \rho'_{lp,l'p'} \tag{5}$$

where λ' is the density matrix of the initial state, ρ' the reduced scattering density matrix and G a geometrical factor:

$$\lambda'_{lm,l'm'}(n; e) = c_{nlm}(e) c^*_{nl'm'}(e)$$

$$\rho'_{lp,l'p'} = \sum_f 2\pi \int_0^\infty b\, \mathrm{d}b\, a'_{f,lp}(b) a'^*_{f,l'p'}(b) \tag{6}$$

$$G(lp, l'p', m, m'; \varphi) = \sum_q (-1)^{p+p'} \mathrm{i}^{m-m'} d^{(l)}_{pq}(\tfrac{1}{2}\pi)\, d^{(l')}_{p'q}(\tfrac{1}{2}\pi)\, d^{(l)}_{qm}(\varphi)\, d^{(l')}_{qm'}(\varphi).$$

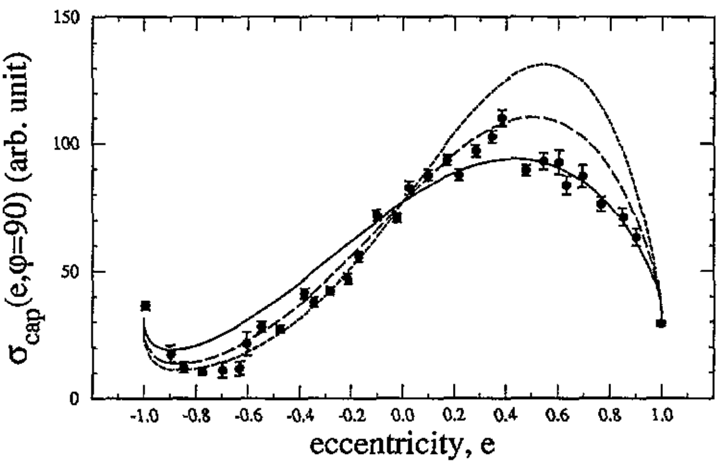

Figure 2. Dependence of electron capture cross sections on the eccentricity of the initial elliptic states where the plane of the ellipse is parallel to the beam direction. The experimental data are from the measurement by Ehrenreich *et al* (1994) for collisions of the Li ($n = 25$) elliptic state by Na^+. The calculations are results from the close-coupling methods for $H^+ + H(n)$ where the initial state $H(n)$ is an elliptic state in the n=3 (full curve), n=4 (long-broken curves) and $n = 5$ (short-broken curves). The relative collision velocity in all cases is 1.6. The data are normalized to each other at $e = 0$.

We have performed standard close-coupling calculations (Fritsch and Lin 1991, Toshima and Eichler 1992) for p+H ($n_i = 3, 4, 5$) collisions at the reduced velocity (ratio of projectile velocity V to the average electron velocity, $\sqrt{\langle v_e^2 \rangle}$) of 1.6. For each system the basis set in the close-coupling calculations consists of all the $n = 1, \ldots, n_i + 1$ states on both centres. In figure 2 we compare the experimental cross section measured as function of e for the Na^+ + Li ($n = 25$, e) system (Ehrenreich *et al* 1994) with the theoretical cross section for the above three systems for φ=90°, i.e. when the beam is parallel to the plane of the orbital. The eccentricity has been generalized to take on both positive as well as negative values, with the convention that for a positive e the classical motion of the electron is along the beam direction at the perihelion, while the opposite is true for a negative e.

The calculated theoretical cross sections have been normalized to the experimental data at e=0 (to obtain the absolute cross sections in units of 10^{-16} cm^2 the $n = 3$, 4 and 5 curve must be multiplied by 0.54, 1.5 and 3.1 respectively). It is clear from the figure that the strong dependence on e for the low n elliptic states of hydrogen is similar to what is observed in the experiment on the elliptic Rydberg state of Li ($n = 25$). Classically the cross section, $\sigma_{cap}(n)$, for electron capture from H(n) scales with n^4, thus the classical values of the ratios $\sigma_{43} = \sigma_{cap}(4)/\sigma_{cap}(3)$ and $\sigma_{54} = \sigma_{cap}(5)/\sigma_{cap}(4)$ are 3.2 and 2.4 respectively. Averaging over the whole range of eccentricities we find for the close-coupling calculations too that the ratio σ_{43} (= 2.7) is bigger than σ_{54} (= 2.1). However, it is not clear that the classical scaling law should work for these small n, and a conclusion about whether this limited series of close-coupling calculations converges towards a universal $\sigma_{cap}(n)$ curve or not, cannot be drawn. Also, one should keep in mind that, mainly due to the limited basis set, the close-coupling calculations are not exact.

Figure 2 shows that for a fast projectile (reduced velocity = 1.6) electron capture from an elliptic state in which the electron is moving along the beam direction at perihelion (positive e) is favoured compared to the reverse orbiting direction of the electron (negative e). Since the classical perihelion velocity of the electron is always bigger than or equal to the average velocity, we interpret the observed e dependence of σ_{cap} in terms of velocity matching in the sense that the perihelion velocity roughly must match the beam velocity. This simple velocity matching interpretation can be tested by calculating $\sigma_{cap}(e)$ for different angles of orientation and by changing the reduced velocity to a value less than one, in which case we have velocity matching at the aphelion. Since, as might be noted from figure 2, the relative capture cross section depends only weakly on the principal quantum number n, we consider only the $n = 4$ case in the following analysis.

The capture cross section, $\sigma_{cap}(e)$, for the p + H ($n = 4$, e) system has been plotted in figure 3 for four different orientation angles, φ=0°, 30°, 60° and 90°. We see that the e-dependence becomes less prominent as we turn the plane of orbit from being parallel to the beam axis (φ=90°) to being perpendicular to the beam axis (φ=0°). In the perpendicular case there is no preferred orbiting direction of the electron because of the symmetry. Also we notice that the peak position of $\sigma_{cap}(e)$ shifts towards larger eccentricities as φ is decreased. This behaviour is in agreement with the velocity matching picture, since the the component of the velocity at the perihelion in the direction of the beam is proportional to $\sin\varphi$. For a tilted ellipse, in order to have velocity matching the velocity at perihelion has to increase or equivalently the eccentricity has to increase.

More specifically, according to classical physics, the component of the perihelion velocity in the beam direction $v_{ep}(e, \varphi)$ is given by

$$v_{ep}(e, \varphi) = \sqrt{\frac{1+e}{1-e} \langle v_e^2 \rangle} \sin\varphi. \tag{7}$$

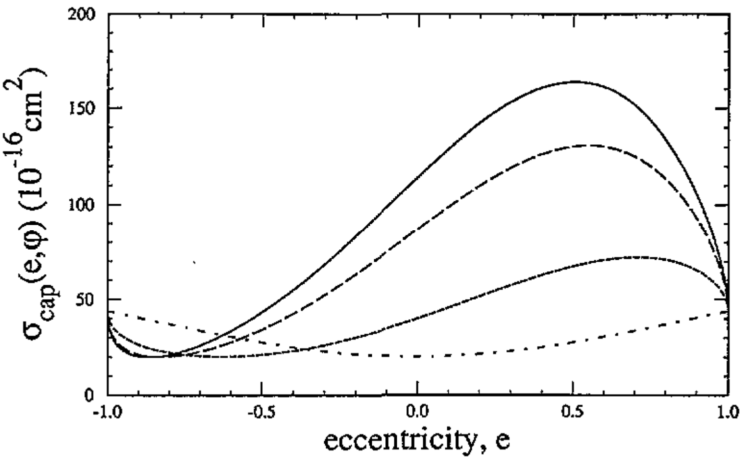

Figure 3. Dependence of electron capture cross sections on the eccentricity for protons colliding with the H ($n = 4$) elliptic states for four different orientation angles of the ellipse: $\varphi = 90°$ (full); 60° (long-broken curves); 30° (short-broken curves); and 0° (chain curves). The results are from the close-coupling calculations for a relative velocity of 1.6.

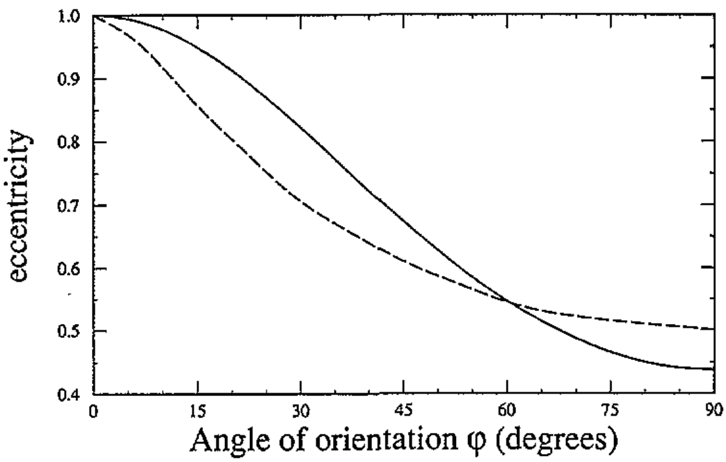

Figure 4. The relation between the eccentricity where the electron capture cross sections peaks versus the orientation angle φ. The full curves are from the close-coupling calculations (part of which are shown in figure 3) and the broken curves are from the classical results based on equation (8).

If we require that this velocity match the beam velocity the eccentricity will be given by v_r and φ

$$e = \frac{v_r^2 - \sin^2\varphi}{v_r^2 + \sin^2\varphi}. \tag{8}$$

In figure 4 we plot the values of the eccentricity where the cross section peaks for different orientation angles φ obtained from the calculation and compare the results with those given by equation (8). Not only does the classical interpretation give the correct φ dependence

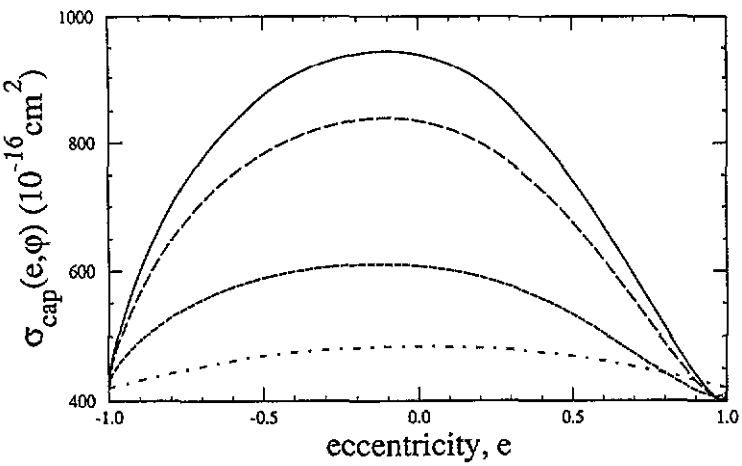

Figure 5. Similar to figure 3 except for collisions at a relative velocity of 0.8.

qualitatively, but also the correct position within about 20%. This indicates that the velocity matching plays a major role in the capture process.

If the reduced collision velocity is less than one, then the velocity matching can occur only at aphelion. Therefore the peak of the capture cross section should occur at a negative eccentricity if the velocity matching is a good criterion. In figure 5 we show how the electron capture cross sections depend on the eccentricity for different angles of orientation φ for a reduced collision velocity of 0.8. In agreement with the criterion of velocity matching the peak position is located at negative eccentricities, although the peak is not as striking indicating that the velocity matching criterion is not as strong for collisions at lower velocities.

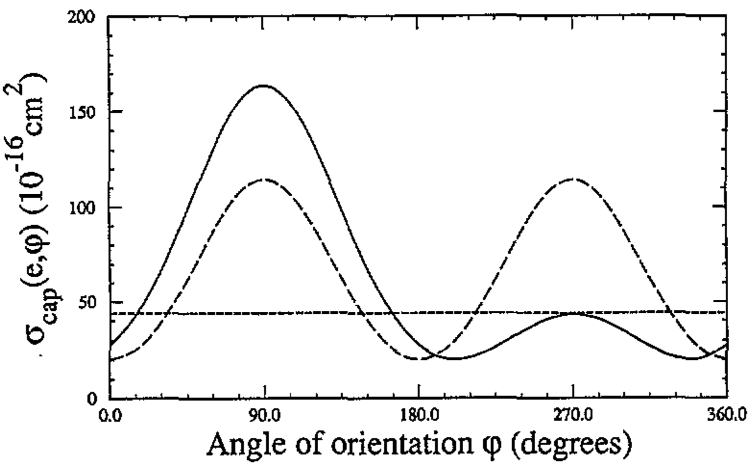

Figure 6. The dependence of electron capture cross sections on the angle of orientation φ for three different elliptic orbitals, $e = 0.5$ (full curve); 0.0 (long-broken curves) and 1.0 (short-broken curves).

So far we have kept φ fixed and only varied e, but it is also of interest to plot σ_{cap} as function of φ and keep e fixed. Figure 6 shows $\sigma_{\text{cap}}(e, \varphi)$ for the three eccentricities $e = 0$, 0.5, and 1.0 in the case of a reduced velocity of 1.6. We notice that capture in general is much more likely to take place when the projectile velocity and the velocity of the Rydberg electron at perihelion are parallel ($\varphi = 90°$) than when they are perpendicular ($\varphi = 0°$). For the case of a circular Rydberg state this effect has been observed experimentally (Hansen *et al* 1993) (for a comparison between experiment and theory see the classical calculations by Wang and Olson (1994) and the semiclassical calculations by Lundsgaard *et al* (1994)). The present calculation shows that the ratio $\sigma(e, \varphi = 90°)/\sigma(e, \varphi = 0°)$ peaks at an eccentricity e between the two extreme cases $e = 0$ and $e = 1$.

Also, figure 6 is another way to show that for a general elliptic state capture is more favoured when the orientation of the elliptic state is so, that perihelion velocity and beam velocity is parallel ($\varphi = 90°$) as compared to when they are anti-parallel ($\varphi = 270°$). For circular states the capture cross section is independent of the orientation of the electron (the integration over all impact parameter planes removes any effect of the orientation, see figure 1), which is reflected in the symmetry around $\varphi = 180°$ of the $e = 0$ curve in figure 6. For $e = 1$, i.e. $B = 0$, the target being in the extreme Stark state has rotational symmetry about the axis of the applied electric field ($-x$ direction in figure 1), and since the φ angle is a rotation around this axis, the $\sigma_{\text{cap}}(e = 1, \varphi)$ is independent of φ as seen in figure 6.

To conclude, close-coupling calculations for protons colliding with the elliptic states of the hydrogen atom in the $n = 3$, 4 and 5 manifolds have shown that the dependence of electron capture cross section on eccentricity is nearly independent of n. (We mention that the n-independence is expected if the collision is describable classically since the classical hydrogen atom can be scaled with energy E.) The observed dependence of the capture cross section on the eccentricity is found to be in agreement with the criteria of velocity matching for the electron capture process.

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