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LETTER TO THE EDITOR

Final state distributions for electron capture from orientated Rydberg atoms

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Abstract. Final state nlm distributions are studied for electron capture from orientated circular Rydberg atoms. Strong orientation effects are found for all quantum numbers at collision speeds comparable to the electron orbital speed. Predominant population of large m states is observed when the plane of the circular orbit is nearly perpendicular to the incident direction of the projectile. The lm distributions show structures due to quasi-Thomas scattering even at low relative collision velocities.

It is important in many areas of physics to control dynamical pathways in order to delineate physical effects under conditions otherwise unavailable. Orientation of atoms by laser pumping is such a case. For example, even for a modest orientation of the atomic p+ orbital by polarized single-photon excitation from an s state, electron capture has shown a strong left–right asymmetry that provides sensitive information about the dynamics that is missing from unorientated s states (Houver *et al* 1992, Lewartowski and Courbin 1992). Much higher degrees of orientation are now feasible owing to recent developments in experimental preparation of circular Rydberg atoms by laser excitation and cross-field techniques (Delande and Gay 1988, Hare *et al* 1988). A circular Rydberg atom has the maximum angular momentum l_0 for a given principal quantum number n_0 such that $l_0 = n_0 - 1$. (Throughout this letter we adopt the notation of n_0, l_0, m_0 for the initial target state and n, l, m for the final projectile state of the captured electron. m_0 and m are the usual magnetic quantum numbers.)

The electron in a circular Rydberg state moves in a very localized region in phase space. It is semiclassical and the plane of the orbit is well defined spatially. The special properties of circular Rydberg atoms as semiclassical objects have significant implications in many fields, especially for electron capture in atomic collisions. Since the plane of the electron orbit can be placed in any desired orientation along a given quantization axis, its spatial orientation allows continuous selection of the component of the electron orbital speed parallel to the quantization axis. There are two extreme orientations that are most significant, $m_0 = l_0$ and $m_0 = 0$. *In the former orientation, the orbital plane is nearly perpendicular* to the quantization axis. The motion of the electron is confined almost entirely within this plane, and the electron has little parallel velocity component to that of the projectile. On the other hand, for the latter orientation, the quantization axis lies within the plane of the electron orbit. The electron has the maximum parallel velocity component along the projectile direction. The parallel velocity component is important for capture in the regime in which the velocity matching mechanism dominates.

Total capture cross sections were predicted (Kohring *et al* 1983) to depend strongly on the two orientations which control the velocity-matching mechanism. New experimental

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data (Hansen *et al* 1993) are in qualitative agreement with the prediction. However, quantitatively, theory and experiments are at variance (Wang and Olson 1993) for the ratio of $m_0 = l_0$ to $m_0 = 0$ cross sections over a given range of product states detectable in the experiment by Stark field ionization. Detailed analysis of this quantitative difference is unavailable due to the lack of knowledge on the final nlm distributions for capture from orientated circular Rydberg atoms.

We report in this letter a theoretical study of the final projectile state nlm distributions following capture from orientated circular Rydberg states. As a prototype reaction, we choose the collision system $\mathrm{H}^+ + \mathrm{H}(n_0 l_0 m_0)$ with the initial circular Rydberg state of $n_0 = 25, l_0 = 24$. A collision speed of $v_p/v_e = 1.5$ is used in this study where v_p and $v_e = \frac{1}{25}$ au denote the proton speed and electron orbital speed, respectively. The above parameters correspond to conditions that have been realized in current experiments (Hansen *et al* 1993). We shall focus on two orientations, $m_0 = 24$ and $m_0 = 0$, where the incoming proton direction is chosen as the quantization axis.

Our primary aim is to obtain information about and to investigate orientation effects on the nlm distributions. As we will show later, strong orientation effects are found for all quantum numbers. Also, predominant population of non-zero, large m states is observed for the $m_0 = 24$ orientation. In addition, structures observed in the m distribution are attributable to the Thomas capture mechanism. Besides its fundamental interest, understanding of these distributions is also crucial in experimental extraction of partial cross sections by selective field ionization techniques (MacAdam *et al* 1990). Although the scope of this letter is limited to single capture only, the information derived may be useful in other studies as well, including multiple capture processes, astrophysics and plasma physics (Barat and Roncin 1992, Chen and Lin 1992, Stebbings and Dunning 1983).

Previous studies of the final state distributions have been done mostly for unorientated, low-l_0 (elliptic) initial states ($n_0 \sim 30, l_0 \leqslant 2$) (Olson 1980, Becker and MacKellar 1984, Burgdörfer and Dubé 1985, MacAdam *et al* 1990, Pascale *et al* 1990). nl distributions were discussed for circular Rydberg states (Becker and MacKellar 1984) but the states were not orientated and no m distributions were given. High Rydberg states ($n_0 \gg 1$) are well described classically. In particular we note that the relative uncertainties associated with the position r and momentum p of the electron in a circular orbit scale as $\Delta r/\langle r\rangle \simeq \Delta p/\langle p\rangle \simeq 1/\sqrt{n}$, $\Delta r \Delta p \simeq \hbar$ (Bethe and Salpeter 1957) such that the position and momentum can be specified very accurately for high Rydberg states without violating the Heisenberg uncertainty principle. Therefore classical dynamics are expected to be valid.

In this study we adopt the well known classical trajectory Monte Carlo method (Abrines and Percival 1966, Olson and Salop 1977) to simulate the dynamical evolution of the collision system. In this method, the initial conditions of the system are sampled randomly from a microcanonical subensemble. The orientation of the initial target state is achieved by inclining the plane of the electron orbit according to the desired m_0 number. The system is then propagated according to its full three-body classical Hamiltonian. Capture events are recorded at the end of evolution. Classical $n_c l_c m_c$ numbers are related to quantum nlm numbers through standard binning procedures

$$
\begin{aligned}
&((n-1)(n-\tfrac{1}{2})n)^{1/3} \leqslant n_c < (n(n+\tfrac{1}{2})(n+1))^{1/3} \\
&l \leqslant l_c < l+1 \\
&m \leqslant m_c < m+1 \quad (\text{for } m_c \geqslant 0) \qquad m-1 < m_c \leqslant m \quad (\text{for } m_c < 0).
\end{aligned}
$$

The n distributions are shown as percentages in figure 1 for two orientations $m_0 = 24$ and $m_0 = 0$ in collisions of $\mathrm{H}^+ + \mathrm{H}(n_0 = 25, l_0 = 24, m_0)$ at $v_p/v_e = 1.5$. A

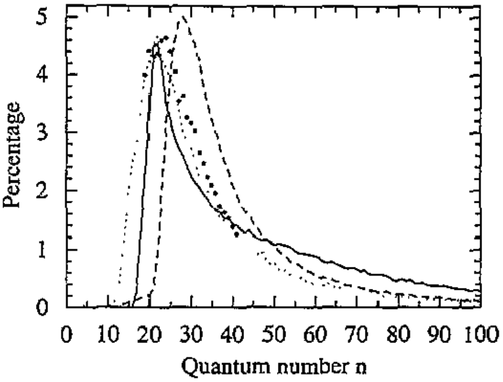

Figure 1. The n distributions, expressed as percentages, for capture in $H^+ + H(n_0 = 25, l_0 = 24, m_0)$ collisions at the ratio of proton to electron orbital speed $v_p/v_e = 1.5$. Full curve, $m_0 = 24$; broken curve, $m_0 = 0$; dotted curve, unorientated initial state $n_0 = 25$, $l_0 = 2$; full circle, experimental data of (Rolfes and MacAdam 1982) for $Na^+ + Na(24d)$ at $v_p/v_e \simeq 1.42$.

calculation for capture from the *unorientated, elliptic* 25d state is plotted in figure 1 as a benchmark. Experimental data (Rolfes and MacAdam 1982) for a slightly different system ($Na^+ + Na(24d)$ at $v_p/v_e \simeq 1.42$) are also included for comparison. The results show general features characteristic of capture from Rydberg states in the velocity matching region, as has been noted before (Olson 1980, Becker and MacKellar 1984, Pascale *et al* 1990, MacAdam *et al* 1990). The n distribution rises rapidly to a peak value centred approximately near n_0 and falls quickly thereafter. The agreement between theory and experiment for unorientated 25d is satisfactory, in view of the slightly different collision systems and corresponding quantum defects for low-l states.

However, large differences exist between the two orientations of the circular state, including the peak positions, the positions for the onset of rapid rise, and their widths. In the $m_0 = 0$ orientation, the electron orbital plane is parallel to the direction of the incoming proton. For approximately half an orbital period, the electron has a positive velocity component parallel to the proton's direction. This effectively lowers the relative collision speed. Therefore, the peak position and the position for the onset of rapid rise are shifted upwards compared to the $m_0 = 24$ orientation. In addition, the slowly decaying tails for large n are also very different. The contributions from $41 \leqslant n \leqslant 100$ account for approximately 43%, 32%, and 24% of the total cross sections for $m_0 = 24$, $m_0 = 0$ and 25d, respectively. It clearly demonstrates that for circular Rydberg states, cross sections arising from large n levels cannot be neglected. It is also important to note that the contributions from large n levels are orientation-dependent.

Figure 2 displays the l distributions for the same three initial states as above. In all three cases, the distributions are relatively broad. Circular initial states $m_0 = 0, 24$ produce narrower distributions than elliptic states as observed previously (Becker and MacKellar 1984). Also for circular states they peak near their initial l_0 value (24 in our case). It is interesting to note that broad l distributions centered at large l-values can be obtained through capture from circular Rydberg states. The only other means of producing such distributions in atomic collisions was via capture in the presence of stochastic forces in ion–solid interactions (Burgdörfer and Bottcher 1988). Doubly or multiply excited states formed by multiple capture by highly charged ions from circular Rydberg atoms may prove important to spectroscopic studies of the atomic three-body problem for systems inaccessible by photon excitations (Eichmann *et al* 1992).

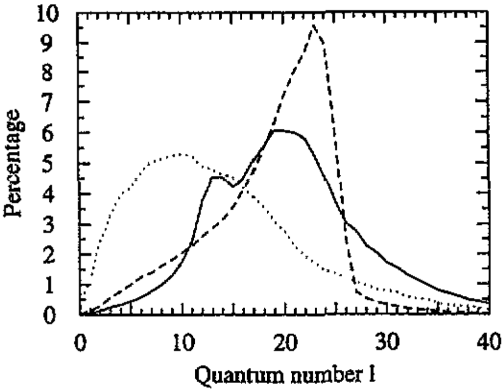

Figure 2. The l distributions for the same collision system as shown in figure 1. Full curve, $m_0 = 24$; broken curve, $m_0 = 0$; dotted curve, unorientated initial state $n_0 = 25$, $l_0 = 2$.

Another striking feature in figure 2 is the occurrence of a secondary peak around $l \sim 14$ in the l distribution for the orientation $m_0 = 24$. Its appearance for this orientation underscores again the importance of orientation effects. The origin of this secondary peak is another signature of the Thomas double scattering mechanism for capture (Thomas 1927) recently discussed in terms of the angular scattering of the proton in collisions with orientated Rydberg atoms (Wang and Olson 1993). In the Thomas capture mechanism, two collisions must occur: the first collision between the electron and the projectile, followed by the second collision between the electron and the target nucleus. The electron is accelerated to approximately the speed of the projectile after the first collision and deflected to the forward direction by the second collision. For the $m_0 = 24$ orientation, direct velocity matching is strongly suppressed due to lack of the electron velocity component parallel to the direction of the proton. This necessitates the first collision. Furthermore, in the plane formed by the electron orbital velocity and the proton velocity, the electron can be scattered to the left or to the right of the proton direction. Because of the non-negligible electron orbital velocity in our case ($v_p/v_e = 1.5$), the momentum transfer is different for left-scattering compared to right-scattering. In particular, left-scattering imparts a large momentum transfer in the opposite direction to the initial electron orbital velocity. The electron acquires certain angular momentum carried into the final state that is reflected in figure 2 as a secondary peak near $l = 14$.

An immediate consequence of the above observation is that the electron is expected to reverse its orbital rotation about the proton direction as well, which should be observable in the corresponding m distribution. We show in figure 3 the m distributions for the three orientations used above. Drastic differences exist among the different initial states and orientations. For both $m_0 = 0$ and 25d initial states, the distributions peak at $m = 0$. This behaviour is expected from reflection symmetry. However, for $m_0 = 24$, the distribution populates overwhelmingly positive-m states, evidenced by the broad peak centred near $m \sim 22$. The distribution shows a 'memory' effect and resembles a diffusion process in m-space from its initial $m_0 = 24$ state. For comparison, the m distribution for $m_0 = -24$ (orientated opposite to $m_0 = 24$ direction) is also displayed in figure 3. It is symmetric with respect to the distribution for $m_0 = 24$ as expected.

It can be seen from figure 3 that there are two additional structures superimposed on the m distribution for $m_0 = 24$ orientation: a peak at $m = -14$ and a shoulder at $m = 24$. They are both related to the left- and right-scattering processes discussed above. The peak

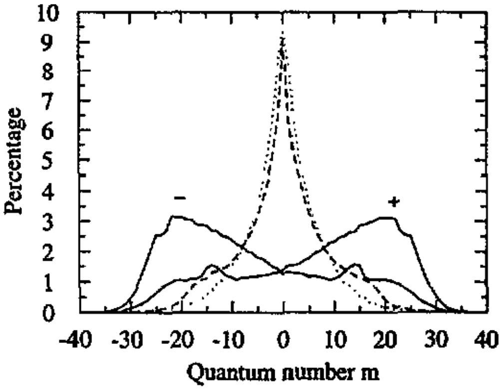

Figure 3. The m distributions for the same collision system as shown in figure 1. Full curves, $m_0 = 24$ (labelled +) and $m_0 = -24$ (labelled −); broken curve, $m_0 = 0$; dotted curve, unorientated initial state $n_0 = 25$, $l_0 = 2$.

near $m = -14$ corresponds to the secondary peak observed in the l distribution (figure 2) and is due to the large momentum transfer imparted in the first collision to the electron in the direction opposite to its initial orbital rotation (m_0). The shoulder structure at $m \simeq 24$ indicates that the right-scattering process has a relatively minor effect on the m distribution because of the smaller momentum transfer. All these features demonstrate that capture mechanisms are sensitively reflected in the l, m distributions. They provide additional evidence for the importance of Thomas capture mechanism to the information derived from the angular scattering of the proton (Wang and Olson 1993).

The m distributions studied here are important for experimental analysis of field ionization of a given range of n levels, as has been pointed out (Pascale *et al* 1990, MacAdam *et al* 1990, Wang and Olson 1993). The couplings of different states of an atom in electric fields are strongly dependent on the m-values of the states (Stebbings and Dunning 1983). Ionization thresholds differ significantly for strongly-mixing states (adiabatic ionization) and weakly-mixing states (diabatic ionization). Experimental determination of specific n-levels is crucial to studying orientation effects. As we have shown (figure 3), the m distributions are completely different for orientations ranging from $m_0 = 24$ to $m_0 = 0$. To accurately determine relative contributions of many n levels by field ionization techniques, a modelling by combining the Stark map under experimental conditions and certain *a priori* m distributions (such as these presented in this study) may prove necessary and fruitful.

In conclusion, we have presented a theoretical study on the final state nlm distributions for capture from orientated circular Rydberg atoms. The nlm distributions depend sensitively on the orientation of the atom. Structures are found in l, m distributions that reveal signatures of the capture mechanisms. The $m_0 = 24$ orientation appears most interesting because of its unusual l- and m distributions. These distributions are important to a number of problems, including renewed interest in cuspology, convoy electron production, and experimental analysis of n levels by field ionization. It may be possible to experimentally test the n, l, m distributions by line emission or electron spectroscopy. The latter is especially promising since it is known that the low-energy electron emission from a Rydberg state has an ellipsoidal angular distribution characterized by a set of multipoles that depend sensitively on the state quantum numbers.

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