

Electron capture from circular Rydberg atoms

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Electron capture cross sections from circular Rydberg states as a function of the angle φ between the ion velocity and the angular momentum of the circular orbital have been reported recently by Hansen *et al.* [Phys. Rev. Lett. **71**, 1522 (1993)]. We show that the observed φ dependence can be explained in terms of the propensity rule that governs the dependence of electron capture cross sections on the magnetic quantum numbers of the initial excited states. We also carried out close-coupling calculations to show that electron capture from the circular H(3*d*, 4*f*, 5*g*) states by protons at the same scaled velocity has nearly the same φ dependence.

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In a recent paper [1] Hansen *et al.* reported electron capture cross sections from circular Rydberg states as a function of the angle φ between the ion velocity and the angular momentum of the circular orbital. They studied the collision of 2.5 keV $^{23}\text{Na}^+$ on the Rydberg state of Li with $n=25$, $l=n-1$ and $m=+l$, where m is defined relative to a weak, external magnetic field. It was found that the electron capture cross section, $\sigma_{cap}(\varphi)$, depends strongly on the angle φ , with the maximum occurring at $\varphi=90^\circ$ and decreases rapidly with decreasing φ (see the experimental data reproduced in Fig. 2 below). The experimental result shows that it is 4.5 times more likely to capture an electron if the circular orbit lies along the beam direction than if the orbit lies perpendicular to it.

Attempts have been made by Hansen *et al.* to interpret this φ dependence in terms of higher-order perturbation theories [2] and in terms of results obtained for lower- n states ($n=10$) calculated using the classical trajectory Monte Carlo (CTMC) model [3]. CTMC calculations for initial circular states have recently been reported by [4] Wang and Olson, who qualitatively find the same dependence on φ of the total capture cross section as observed in the experiment by Hansen *et al.*, but overestimate the ratio $\sigma_{cap}(\varphi=90^\circ)/\sigma_{cap}(\varphi=0)$ by a factor of 3. In this paper, we show that the observed φ dependence is consistent with the known propensity rule for electron capture from low-lying excited states. This propensity rule has been proved based on results from elaborate close-coupling calculations [5,6] and from experiments by protons on oriented Na(3*p*) states [7]. We also carried out close-coupling calculations to determine the φ dependence for electron capture from the circular H(3*d*, 4*f*, 5*g*) states by protons at the same scaled collision velocity (the ratio of the projectile velocity to the initial orbital velocity of the electron), and showed that the calculated φ dependence is nearly identical to the measured result for the $n=25$ circular state by Hansen *et al.*

To specify the collision more precisely, we first define the "natural frame" of reference, see Ref. [8]. In this frame the collision plane is the $x'y'$ halfplane where y' is posi-

tive and $+x'$ is the direction of the incident beam. The quantization axis ($+z'$ direction) is perpendicular to the scattering plane such that $x'y'z'$ forms a right-handed coordinate system (see Fig. 1). A circular Rydberg state $|i\rangle_B = |n, l=n-1, m_i=l\rangle_B$, where m_i is given with respect to the z_B axis (see Fig. 1) can be obtained from rotating the circular state $|n, l=n-1, m_i=l\rangle'$ defined with respect to the natural frame $x'y'z'$ by two successive rotations,

$$\begin{aligned} |i\rangle_B &= D(\eta + \pi/2, \pi/2, \pi) D(0, -\varphi, 0) |nlm_i\rangle' \\ &= \sum_q \sum_m \mathcal{D}_{qm_i}^{(l)}(0, -\varphi, 0) \\ &\quad \times \mathcal{D}_{mq}^{(l)}(\eta + \pi/2, \pi/2, \pi) |nlm\rangle'. \end{aligned} \quad (1)$$

In the equation above, D is a rotation operator and \mathcal{D} the corresponding rotation matrix defined as in Edmonds [9]. The angles are defined in Fig. 1, where η is the rotation angle of the scattering plane $x'y'$ with respect

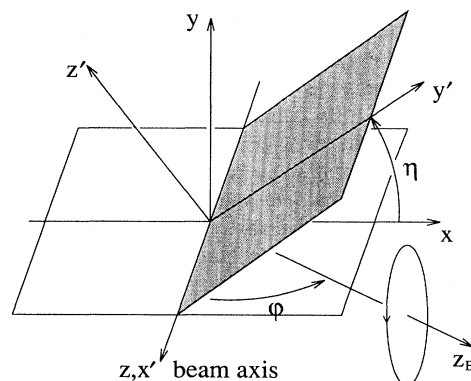


FIG. 1. The geometry for describing the collisions. The ion beam is traveling in the direction of z (and x'). The xyz is a space-fixed frame of reference. The scattering plane is the $x'y'$ plane which makes an angle η with respect to the space-fixed xz plane. The z_B is the direction where the circular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

to the space fixed xz plane ($z = x'$). The amplitude $a_{fi}^B(\varphi, b, \eta)$ for the transition $|i\rangle_B \rightarrow |f\rangle$ depends on the angles φ, η , and the impact parameter b . This amplitude can be expressed as a coherent sum of amplitudes defined with respect to the natural frame,

$$a_{fi}^B(\varphi, b, \eta) = \sum_q \sum_m i^q (-1)^m \exp(iq\eta) d_{mq}^{(l)}(\pi/2) \times d_{m,q}^{(l)}(\varphi) a'_{fm}(b). \quad (2)$$

By integrating the transition probabilities over the impact parameter plane and summing over all the final states, an expression for the total electron capture cross section from a circular state ($m_i = l$) oriented in the direction of z_B can be obtained,

$$\sigma_{cap}(\varphi) = \sum_q [d_{m,q}^{(l)}(\varphi)]^2 \times \sum_m \sum_{m'} d_{mq}^{(l)}(\pi/2) d_{m',q}^{(l)}(\pi/2) \rho'_{mm'}, \quad (3)$$

where

$$\rho'_{mm'} = \sum_f 2\pi \int_0^\infty a'_{fm}(b) a'_{f m'}^*(b) b db. \quad (4)$$

Since the symmetry of reflection with respect to the collision plane is a conserved quantity, an initial state with even (odd) symmetry will only couple to even (odd) final states. This implies that $\rho'_{mm'} = 0$ unless $m + m'$ is an even number. Equation (3) gives the total electron capture cross section for a particular magnetic substate (m_i) where the quantization axis is along the z_B direction. This cross section is expressed as a coherent sum over the integral density matrices (4) where the latter are defined with respect to a fixed "natural frame," weighted properly by the rotational matrix elements. For electron capture from a Rydberg state, the full knowledge of the density matrix is hard to obtain. However, the φ dependence of (3) can be estimated based on the propensity rule that describes the relative magnitudes of $\rho'_{m,m}$ where m is defined with respect to the natural frame [5–8].

According to the propensity rule for electron capture from excited states, the probability is largest for $m = -l$ for a given (n, l) and decreases rapidly with increasing values of m . This result is valid only if the quantization axis is perpendicular to the scattering plane as defined in the natural frame. It implies that electron capture is more likely if the electron is initially rotating in the same sense as the rotation of the internuclear axis (negative m) and if the electron lies close to the scattering plane ($|m| = l$). The propensity rule has been shown to be valid when the ion velocity is close to the orbital velocity of the electron in the initial state. This latter condition is satisfied in the experiment of Hansen *et al.*

We first assume that in the natural frame the cross section for electron capture is nonzero only if the initial $m = -l$. By setting $m = m' = -l$ in Eq. (3), the electron capture cross section from a circular state oriented in the z_B direction becomes

$$\sigma_{cap} = \rho'_{-l,-l} \sum_q [d_{lq}^{(l)}(\varphi) d_{-lq}^{(l)}(\pi/2)]^2. \quad (5)$$

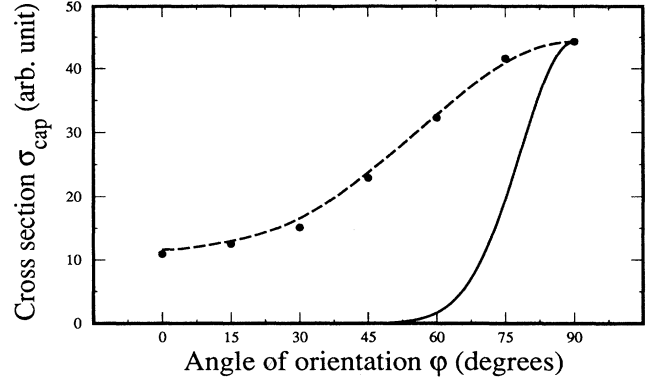


FIG. 2. The φ dependence of the total electron capture cross section from a circular Rydberg state ($n=25, l=24, m=24$) measured by Hansen *et al.* The two curves are from theoretical models (see text). The scaled velocity is 1.66.

Under this approximation, the resulting φ dependence is shown as the solid line in Fig. 2 where the cross section is normalized to the experimental data of [1] at $\varphi = 90^\circ$. The predicted φ dependence has the correct trend, but it decreases too rapidly with decreasing angle φ as compared to experimental data.

To achieve better agreement with experiment, it is necessary to include other m and m' terms in Eq. (3). Without actual calculations available for the whole density matrix $\rho'_{m,m'}$, we will make an estimate first by neglecting the off-diagonal terms and approximate (3) as

$$\sigma_{cap}(\varphi) = \sum_m G_m(\varphi) \rho'_{mm}, \quad (6)$$

$$G_m(\varphi) = \sum_q [d_{m,q}^{(l)}(\varphi) d_{mq}^{(l)}(\pi/2)]^2.$$

First we notice that the maximum point of the geometrical factor $G_m(\varphi)$ is shifted towards smaller φ angles as m is increased from $-l$ to 0, as illustrated in Fig. 3. In fact, if $m = 0$, the maximum occurs at $\varphi = 0$. This result

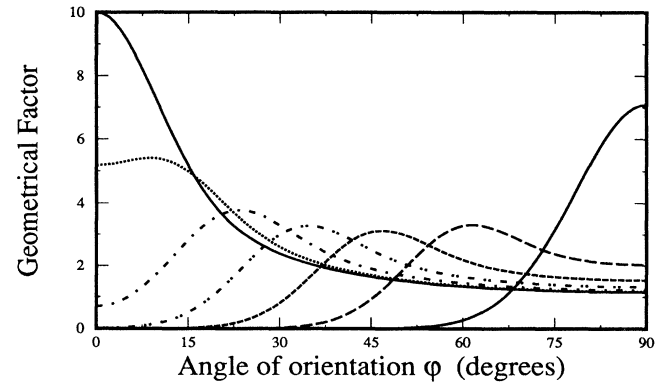


FIG. 3. The geometrical factor $G_m(\varphi)$ of Eq. (6), where m refers to the quantization axis in the natural frame. The solid curve with the maximum at 90° is for $m = -24$. The successive curves with peaks at decreasing φ are for $m = -20, -16, \dots$, respectively. The solid curve with peak at $\varphi = 0$ is for $m = 0$.

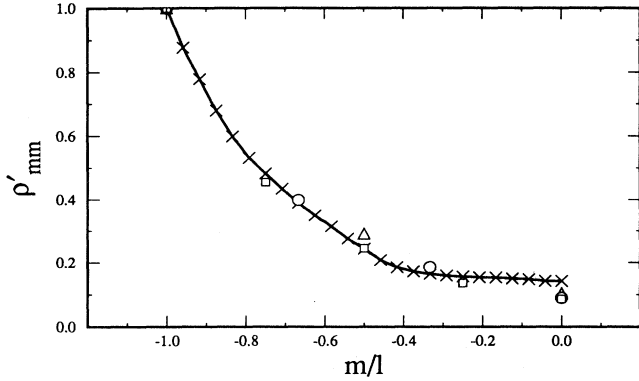


FIG. 4. The diagonal elements of the integral density matrix ρ'_{mm} , as a function of the scaled m -quantum number m/l . Crosses and solid line, visually fitted values for the $\text{Na}^+ + \text{Li}(n=25, l=24, m=24)$ system; open triangles, close-coupling calculations for $p+\text{H}(3d, m=2)$; open circles, close-coupling calculations for $p+\text{H}(4f, m=3)$; open squares, close-coupling calculations for $p+\text{H}(5g, m=4)$.

shows that the $m \neq -l$ terms in Eq. (3) are expected to bring the theoretical calculations at small angles into better agreement with the experimental data.

To make a prediction on the φ dependence based on Eq. (6), we need to make a model for $\rho'_{m,m}$. Because of the propensity rule, we assume that all the positive m do not contribute to the sum. Thus we need only to make a model for the relative values of $\rho'_{m,m}$ for $m = -l, -l+1, \dots, 0$ which should decrease monotonically with increasing values of m . By a simple visual fitting, we found that the experimental φ dependence can be well reproduced (shown as the dashed line in Fig. 2) if the relative values of $\rho'_{m,m}$ are given as shown in Fig. 4.

The validity of the fitted values for $\rho'_{m,m}$, as shown in Fig. 4, can be checked only if actual numerical calculations are available. This is not likely to happen in the near future for electron capture from Rydberg states. An alternative is to examine the φ dependence for electron capture from low-lying circular states. We thus carried out close-coupling calculations for electron capture cross sections for protons colliding with $\text{H}(3d, 4f, 5g)$ at the same scaled velocity (details of the calculations are given in [5]) as in the experiment of Hansen *et al.* We then used Eq. (3) to obtain the φ dependence for electron capture from such circular states. The results for $\text{H}(4f)$ are shown in Fig. 5, together with the data of Hansen *et al.* Interestingly, the agreement between the calculations for the circular state for $n = 4$ and the experimental results for the circular state for $n = 25$ is amazingly good. The capture cross sections from $\text{H}(3d)$ and $\text{H}(5g)$ show the same general φ dependence, but the ratio $\sigma(\varphi = 90)/\sigma(\varphi = 0)$ is ~ 3 in the $\text{H}(3d)$ and ~ 9 in the $\text{H}(5g)$ case compared to ~ 5 for the $\text{H}(4f)$ calculation. The variation may be attributed to the lack of full convergence in the close-coupling calculations. However, the calculations show that the qualitatively dependence of the capture cross section on φ is not very sensitive to the principal quantum number of the initial Rydberg state (classical theory predicts identical dependence). This in-

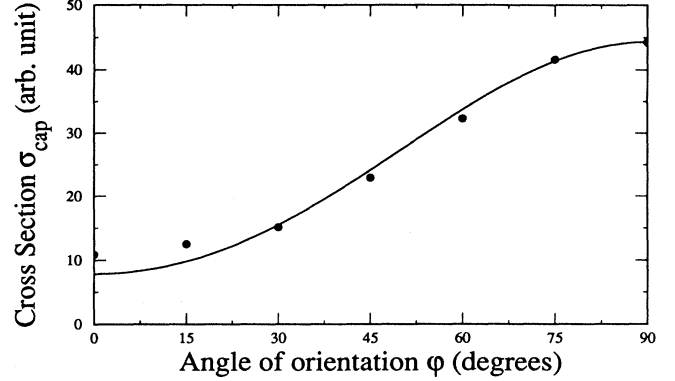


FIG. 5. Comparison between the experimental cross section for electron capture from a circular Rydberg state ($n=25, l=24, m=24$) and the theoretical cross section for capture from a ($n=4, l=3, m=3$) circular state. The scaled velocity is 1.66 in both cases. Dots, Hansen *et al.*; solid line, present calculations.

sensitivity with respect to n can be further illustrated by comparing the diagonal elements $\rho'_{m,m}$ calculated for $n=3, 4$, and 5 with those obtained by “fitting” the experimental data. In Fig. 4 we show in open triangles, circles, and squares the relative $\rho'_{m,m}$ actually calculated for $n=3, 4$, and 5, respectively. By plotting the relative $\rho'_{m,m}$ with respect to the ratio of m/l , it appears that it falls into a universal curve for a fixed scaled velocity and varies very little with the principal quantum number n . The fitted curve in Fig. 2 and Fig. 4 might indicate that the off-diagonal terms in Eq. (3) become less important as n is increased.

The effect of neglecting the off-diagonal elements in Eq. (3) has been checked in all the three cases $n=3, 4$, and 5. We find that the cross section is increased for φ less than roughly 55° and decreased at higher values of φ reducing the ratio $\sigma_{cap}(\varphi = 90)/\sigma_{cap}(\varphi = 0)$ to 1.9, 2.3, and 2.6 for $n=3, 4$, and 5, respectively. The general trend, however, still holds. Truncating expression (6) by excluding the terms with positive m reduces the absolute capture cross section obtained from Eq. (6) by a constant amount — 10% on an average — leaving the relative cross section unaltered.

Before we draw the conclusion, it is interesting to note that the mathematical expression for the φ dependence takes the simpler form [1]

$$\sigma_{cap}(\varphi) = \sum_m [d_{m,m}^{(l)}(\varphi)]^2 \rho_{mm}, \quad (7)$$

if the quantization axis is chosen to be along the beam direction. However, the density matrix $\rho_{m,m}$ in this reference frame does not disclose any propensity rule, and a large range of m values contributes to the summation in Eq. (7).

In conclusion we have shown that the observed φ -dependence of electron capture cross section from a circular Rydberg state by Hansen *et al.* can be interpreted in terms of the propensity rule that governs the dependence of total electron capture cross section on the mag-

netic quantum number of the initial excited state. Our analysis shows that the observed φ dependence is an indirect proof of the validity of the propensity rule for electron capture processes. While a full quantal calculation for such an angular dependence is not available for electron capture from circular Rydberg states, we show that nearly the same φ dependence is obtained for electron capture from the circular $3d$, $4f$, and $5g$ states. Our calculations suggest that the φ dependence varies weakly with the principal quantum number of the circular state. Since the propensity rule becomes less valid at lower col-

lision velocities, we expect a weaker φ dependence for such collisions. Preliminary data by Hansen *et al.* seems to be in agreement with this conclusion.

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