Dominance of the Thomas Mechanism for Electron Capture from Orientated Rydberg Atoms

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We investigate electron capture from initially oriented circular Rydberg atoms by proton impact at collision speeds comparable to the electron orbital speed. The Thomas double scattering mechanism is observed even at these low relative speeds. Furthermore, we find evidence for the dominance of a quasi Thomas capture mechanism in the form of not a single peak, but a double-peaked structure in the differential cross section when the plane of the circular orbit is nearly perpendicular to the incident direction of the projectile.

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Recent progress in experimental techniques for preparing Rydberg atoms of large angular momenta by laser excitation shows great promise in the study of various aspects of atomic physics under novel conditions that were unavailable a few years ago. The significance of Rydberg atoms has been realized in many fields [1], including classically chaotic quantal systems [2], spectroscopy and measurement of fundamental constants [3], and structure of three-body Coulomb systems such as the planetary atoms and doubly excited states [4]. The ability to produce circular Rydberg atoms ($l \sim n-1$, n,l being the principal and the angular momentum quantum numbers, respectively) of sufficient target density in any desired orientation using cross fields methods [5,6] affords the opportunity also to investigate the collision dynamics in the semiclassical regime.

From the viewpoint of studying the collision dynamics, electron capture from a circular Rydberg state is particularly attractive for several reasons. The state is semiclassical with localized charge densities. With the "classical" atom, we can study the fundamental issues related to electron capture, most notably, the dominance of the double scattering capture mechanism that was proposed by Thomas [7] in 1927 in the context of classical mechanics but has not been unambiguously observed within the framework of classical dynamics [8]. Although the Thomas mechanism has been established in quantum calculations [9] and experimental measurements [10] of differential cross sections for capture from atomic $1s$ states, it accounts for only a small fraction of the *total* cross sections at high energies, far below the dominance that Thomas envisioned. The answer to the lack of the signature of the Thomas mechanism in classical dynamics and to the nondominant contribution in experimental total cross sections remains an outstanding and sought-after issue related to the three-body problem. Study of this three-body Coulomb problem addresses not only fundamental questions of charge transfer in atomic collisions, but can also lead to improved understanding of interactions in astrophysics and plasma physics.

In this Letter, we report the first theoretical study of differential cross sections and the prediction of a double-peak structure in these angular cross sections for electron capture from oriented circular Rydberg atoms. We investigate capture mechanisms for projectile speeds v_p comparable to or slightly greater than the electron orbital speed v_e, $v_p \geq v_e$; speeds which are easily realizable in benchtop, laboratory experiments. Our goal is to explore capture from oriented Rydberg atoms, where we can finally remove the restrictions that have prohibited the realization of the dominance of the Thomas mechanism for electron capture. Two important factors accentuate such a realization: (a) The dynamical pathways can be controlled by the manipulation of the orientation of the semiclassical atom [11]. The spherical symmetry of the initial state is broken by orientation, so that the average $\hat{\mathbf{z}}$ ($\hat{\mathbf{z}}$ defined as the incoming projectile direction) component of the electron velocity can be controlled; and (b) the initial and final Rydberg states of the electron are describable classically, enhancing the Thomas mechanism which is essentially a classical process. For the first time, we find evidence for the dominance of the Thomas capture mechanism in total cross sections at low relative speeds $v_p/v_e \geq 1.5$.

For electron capture from *spherically symmetric* states at intermediate to high energies ($v_p > v_e$), it is well known [12] that two mechanisms play an important role: direct and indirect velocity matching. Direct velocity matching proceeds through a first order process and can be described theoretically by the Oppenheimer-Brinkman-Kramers (OBK) approximation [13]. As the projectile speed v_p increases, the indirect velocity matching mediated through multiple scattering begins to dominate. In particular, Thomas proposed a double scattering mechanism [7] as the dominant process at high collision speeds $v_p \gg v_e$, in which the electron is first scattered by the impinging projectile at an angle of 60° with respect to the $\hat{\mathbf{z}}$ direction. Subsequently, a second scattering at the target nucleus by another 60° deflects the electron in the forward direction with near zero velocity relative to the projectile. The primary requirement for the first collision between the projectile and the electron is to accelerate the electron to the speed of the projectile. In what follows, we shall refer to the first collision as the *primary*

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collision. As a result of the primary collision, the projectile is scattered to an angle of ~0.47 mrad, known as the Thomas peak. We note, however, that the Thomas capture process from a spherically symmetric initial state gives only a minor contribution to the total capture cross section, typically at a < 10% level. In fact, in order for the double scattering to be even observable for collisions of protons with atomic hydrogen or helium, impact energies of the order 5 MeV/u ($v_p \simeq 14$ a.u.) are required [10]. Rydberg atoms, on the other hand, have been argued [12] as better candidates for the observation of the Thomas process. Indeed, state selective capture from the ground state into Rydberg states [14] has shown signs of the process at relative low speeds.

Circular Rydberg atoms ($l=n-1$) are ideal candidates for the removal of the conditions that suppress the dominance of the double scattering mechanism. We recall that the relative uncertainties associated with the position r and momentum p of the electron in a circular orbit scale as $\Delta r/\langle r\rangle \simeq \Delta p/\langle p\rangle \simeq 1/\sqrt{n}$, $\Delta r \Delta p \simeq \hbar$ [15] such that the position and momentum can be specified very accurately for high Rydberg states ($n \gg 1$) without violating the Heisenberg uncertainty principle. Since both experiment [16] and theory [17] indicate that capture from an initial Rydberg state of the target populates predominantly a final Rydberg state of the projectile, classical dynamics is expected to be valid throughout the collision.

In the following, we consider capture in the prototype $H^+ + H(nlm)$ collisions within the framework of classical dynamics. For our study, we shall focus on the circular state $n=25$, $l=n-1=24$. Two cases of the magnetic quantum number will be studied in detail, $m=0$ and $m=l=24$. They correspond to the electron orbital plane oriented approximately parallel and perpendicular, respectively, to the incoming proton direction that is defined as the axis of quantization $\hat{z}$. We shall refer to the angle formed between the normal of the orbital plane and the $\hat{z}$ axis as the orientation angle.

In order to simulate the dynamical evolution of the collision system, we adopt here the well-known classical trajectory Monte Carlo method [11,18]. In brief, the initial conditions of the collision system are sampled randomly from a microcanonical subensemble from which l,m are chosen by selecting the appropriate eccentricity and orientation angle of the Kepler orbit. The system is then propagated according to its full three-body classical Hamiltonian. The exit channels are analyzed and capture events are recorded at the end of the evolution when the free particles are sufficiently far apart. Total and differential cross sections are determined by the number of capture events and the impact parameter range.

The total cross sections as a function of the continuous orientation angle are displayed in Fig. 1 for four reduced speeds $v^* = v_p/v_e$. The angular range [0°,90°] corresponds approximately to the magnetic substates from $m=24$ to $m=0$. There is little angular dependence for $v^*=1$. For $v^* > 1$, both global and local changes can be observed. In the $m=0$ orientation, there is sufficient high-momentum component along $\hat{z}$ for direct OBK capture. On the other hand, when the parallel component of the electron orbital velocity is strongly suppressed due to orientation ($m=24$), higher order interactions are required to mediate capture, resulting in much smaller cross sections.

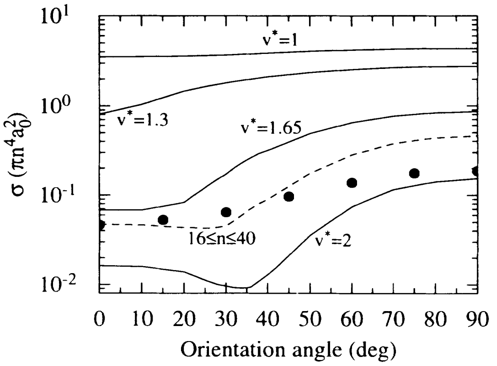

FIG. 1. Total cross sections, in units of $\pi n^4 a_0^2$ with a_0 being the Bohr radius, for capture from $H^+ + H(n=25, l=24, m=0\text{–}24)$ for four reduced speeds $v^* = v_p/v_e$ as a function of the orientation angle. Full curves: theory results; dashed curve: partial capture cross section into final states of $16 \le n \le 40$ for $v^* = 1.65$; full circle: relative experimental data of Ref. [19] corresponding to the conditions for the dashed curve, normalized to theory at 0°.

Very recent experimental data [19] have become available for capture at $v^*=1.65$. They are shown in Fig. 1. We find qualitative agreement between experiment and theory. Quantitatively, theory shows a ratio of $\sigma(90°)/\sigma(0°) \sim 12$, while experiment has a ratio of ~4. In the experiment Stark ionization methods are used to detect product states $16 \le n \le 40$ under the assumption that a given n manifold is uniformly populated [20]. We have also calculated the cross sections by including only these levels. With this modification it appears remarkable that the position for the onset of the rapid rise is about the same in both theory and experiment, both at ~30°. However, the absolute cross sections are reduced by approximately a factor of 2 due to loss of flux from $n > 40$, with the $\sigma(90°)/\sigma(0°)$ ratio essentially unchanged. Although the experiment was conducted with the collision system $Na^+ + Li$, the quantum defect for $l=24$ is negligible, making the use of $H^+ + H$ in theory a satisfactory approximation. Moreover, we note that capture from oriented circular states ($l \simeq n-1$) produces different n,l,m distributions than from elliptic states ($l \sim 0\text{–}2$). These distributions affect the final m levels of the captured electron, which in turn influences the band of n levels experimentally detected by Stark ionization yielding noticeable differences in the observed cross sections [17,20]. A detailed comparison would require a modeling

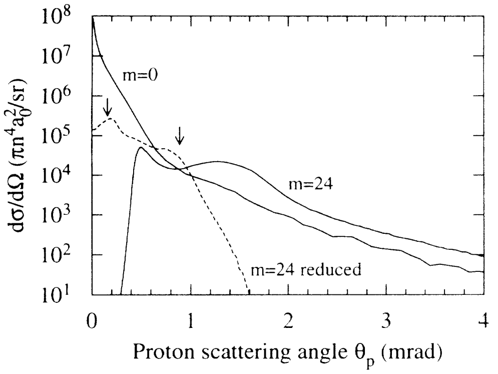

FIG. 2. Differential cross section for capture from $H^+ + H(n=25, l=24, m=0,24)$ as a function of the proton scattering angle for $v_p/v_e = 1.5$. Full curves: results of full three-body simulation for $m=0$ and $m=24$, respectively; dashed curve: reduced three-body simulation by switching off the internuclear interaction for $m=24$ (see text). The arrows mark the positions of the proton deflection angles in a binary collision with the electron using two-body kinematics (see text).

of Stark ionization by combining the Stark map under the experimental conditions and certain *a priori m* distributions.

Figure 2 displays the capture cross section differential in proton scattering angle θ_p for $m=24$ and $m=0$ at a reduced speed of $v^* = 1.5$. The behavior of the cross sections for the two orientations at small proton scattering angles ($\theta_p < 0.2$ mrad) sharply contrasts each other: One ($m=0$) peaks at zero scattering angle as expected from direct velocity matching, while the other ($m=24$) has negligible intensity in this region. Even more remarkable is the fact that, instead of a single peak, there are two peaks in the latter case at rather large angles. The two peaks can be traced to the quasi Thomas double scattering mechanism by noting that at $v^* = 1.5$, one cannot neglect the electron orbital speed as is done in the original Thomas mechanism.

For simplicity, let us assume that the orbital plane is exactly perpendicular to the $\hat{\mathbf{z}}$ direction, so that the electron orbital velocity is perpendicular to the z axis as well. Two-body kinematics for the primary collision between the proton and the electron yields

$$\theta_p = (v_e' \sin\theta_e \pm v_e)/M_p v_p \,, \tag{1}$$

where θ_p is the proton scattering angle in radians ($M_p = 1836$ a.u. is the proton mass), and to zeroth order in $1/M_p$

$$v_e' = v_p \cos\theta_e + \sqrt{v_p^2 \cos^2\theta_e + v_e^2} \,. \tag{2}$$

Equation (2) expresses the magnitude of the scattered electron velocity v_e' in terms of its scattering angle θ_e. We note that because of the finite, non-negligible electron speed, there are two proton scattering angles that depend on whether the electron scatters to the right or the left of the proton. (Here the right-left direction is defined with respect to the $\hat{\mathbf{z}}$ axis in the plane formed by the projectile speed and the electron orbital speed.) The scattering angle of the electron θ_e may be determined by the capture requirement $v_e' \simeq v_p$. Combining Eqs. (1) and (2), the values of θ_p in this case ($v^* = 1.5$) are predicted to be 0.16 and 0.89 mrad, respectively. The two peak angles appearing in Fig. 2 (0.5 and 1.2 mrad, respectively) are obviously much larger than the predictions. We must, however, keep in mind that the internuclear Coulomb interaction also influences the angular scattering due to the small laboratory projectile speed, which is only 0.06 a.u.

To test this effect, we have simulated the capture process in a reduced system by only switching off the nuclear-nuclear interaction. The results are shown in Fig. 2. The two peaks shift dramatically downward to their new positions 0.19 and 0.72 mrad, respectively. The new values are in good agreement with the predictions of the two-body kinematics. This observation is consistent with the model proposed by Fermi [21] who argues that the separation between the Rydberg electron and the target nucleus is so large that they do not interact with the projectile simultaneously. Furthermore, the slowly decreasing Coulomb tail seen in the full three-body calculation is absent in the reduced system as expected. However, the total cross sections of the three-body system and of the reduced system are almost identical, within 0.6% of each other. It reaffirms the assumption [13] that the internuclear interaction does not affect appreciably the total capture cross sections. It is also worth noting that in the limiting case $v_p/v_e \gg 1$, the well-known binary encounter law $v_e' = 2v_p \cos\theta_e$ is recovered from Eq. (2). By requiring $v_e' = v_p$ for capture, we obtain an angle of 60°, which leads to the famous Thomas angle $\theta_p = (\sin 60°)/M_p$.

From the discussions above, we arrive at the following physical picture for capture. For the orientation ($m=24$) nearly perpendicular to the incoming proton direction, direct velocity matching is strongly suppressed due to lack of parallel component of the electron velocity. In order for capture to occur, the primary collision between the electron and the proton must take place in order to accelerate the electron to the speed of the proton. After the primary collision, the electron must also scatter at the target nucleus in order to be deflected to the forward direction, thus completing the quasi Thomas process. The signature of the primary collision appears as two peaks in the differential cross section due to the finite electron orbital velocity. It is clear from Fig. 2 that the quasi Thomas mechanism dominates the total cross section.

Another question arises as to whether the dominance can be observed for a spherically symmetric circular Rydberg atom. We show in Fig. 3 the ratio of the total cross sections for the two orientations ($m=0,24$) as a function of the reduced speed v^*. Initially the ratio decreases for increasing v^*. In this phase, direct velocity matching is dominant over double scattering. As v^* increases above

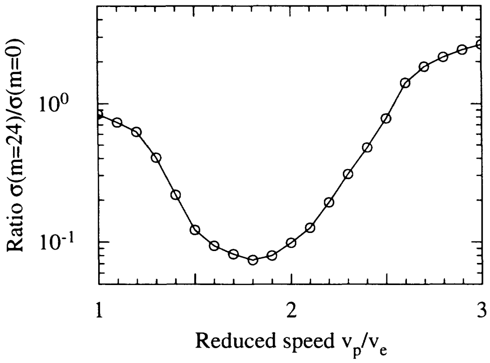

FIG. 3. The ratio of the total cross sections $\sigma(m=24)$ and $\sigma(m=0)$ for capture from $H^+ + H(n=25, l=24)$ as a function of the reduced speed v_p/v_e.

~ 1.8, the ratio starts to increase, indicating the increasing importance of double scattering relative to direct velocity matching. At $v^* \sim 2.5$, the quasi Thomas double scattering reaches a level comparable to direct velocity matching. Above $v^* = 2.5$, Thomas capture overtakes direct velocity matching to become the dominant mechanism. Even at $v^* = 2.5$, capture from $m=0$ orientation is expected to contain a mixture of direct velocity matching, and, to a lesser extent, double scattering. The latter will eventually dominate over the former at sufficiently large v^* for $m=0$ orientation as well. Therefore, we find that at speeds $v^* \geq 2.5$, the dominance of the double scattering will finally be realized for unoriented, *spherically symmetric* circular Rydberg atoms.

In conclusion, we have performed a theoretical study on capture mechanisms from oriented circular Rydberg atoms. For the first time, we have found theoretical evidence of the dominance of the Thomas double scattering from a well-defined initial state in an experimentally reliable energy regime. The signature exists in the form of a double-peak structure in the differential cross section. Our results are general, owing to classical scaling invariances and the correspondence principle. It is expected that experimental data will be available in the near future for a direct comparison of the differential cross sections. Among many other exciting studies that can be explored using circular Rydberg atoms, two deserve special mention: the differential cross sections for capture at large impact speeds and large n, and the ionization spectra of the ejected electrons. The former is important in establishing the dominance of the Thomas mechanism for unoriented Rydberg atoms, while the latter is important in understanding the convoy electrons resulting from ion-solid interactions.

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