Ion Collisions with Rydberg Atoms in Strong Electric Fields

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The classical-trajectory Monte Carlo method has been used to investigate collisions of ions and Rydberg atoms in strong dc electric fields. Cross sections are presented for $n = 10$ and $n = 20$ Rydberg atoms at velocities $1 \leq v/v_e \leq 10$ where $v_e = n^{-1}$ a.u. Electric fields which ionize product Rydberg atoms in states $n' = n + \Delta n$ with $\Delta n = 1$, 2, and 4 were used. The electric field caused the cross sections for electron capture to decrease by up to fourfold while the ionization values increased by up to two orders of magnitude.

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Collisions of ions with Rydberg atoms is a field of active experimental[1-4] and theoretical investigations.[5,6] Besides offering the possibility of the study of weakly bound systems, Rydberg atoms offer a unique advantage in the study of collision dynamics in strong electric fields. This arises because the field strengths required to ionize a Rydberg atom vary as $\sim n^{-4}$, where n is the principal quantum number of the Rydberg atom.[7] As an example, a Na Rydberg atom in the $n = 20$ state requires a field of only ~ 2 kV/cm to ionize the atom[8]; such a field is readily obtained in the laboratory. Further interest in these collision processes arises in the application of Rydberg atoms in far-infrared photon detectors.[9-11] Here, the detection principle is based on the use of a Rydberg atom in state (n, l) absorbing a photon to induce a transition to state (n', l'), which is then easily detected as an ion by the use of field ionization. Collisional processes which lead to the production of ions in the electric field can be a serious limitation to the application of these detectors. In this Letter, we address the subject of the prediction of some of the trends of the cross sections for ion–Rydberg-atom collisions in strong dc electric fields. Both electron capture and ionization processes are investigated along with possible effects of different field orientations. We know of no other calculations or measurements on this topic.

The theoretical procedure employed in the calculation of the cross sections for electron capture

$$A^+ + B(n) \rightarrow A(n') + B^+ \tag{1}$$

and ionization

$$A^+ + B(n) \rightarrow A^+ + B^+ + e \tag{2}$$

was the three-body, three-dimensional classical-trajectory Monte Carlo method. The details of this method have been given previously.[12] We will only discuss the changes required for this set of computations.

For collision studies in an electric field it is not appropriate to cast the formulation into the center-of-mass system which reduces the number of coupled equations, but to maintain all the Cartesian coordinates of the three particles so that the electric field direction can be directly incorporated. The classical form of the Hamilton then is

$$H = \sum_{i=1}^{3} (p_i^2/2m_a + ez_aE_iq_i) + \sum_{i=4}^{6} (p_i^2/2m_b + ez_bE_{i-3}q_i) + \sum_{i=7}^{9} (p_i^2/2m_c + ez_cE_{i-6}q_i) + V(q_1, q_2, \ldots, q_9) \tag{3}$$

with eighteen coupled first-order differential equations arising from

$$dq_i/dt = \dot{q}_i = \partial H/\partial p_i \tag{4}$$

and

$$dp_i/dt = \dot{p}_i = -\partial H/\partial q_i. \tag{5}$$

In (3), m_a, z_a, m_b, z_b, m_c, and z_c are the masses and charges of the three particles, p_i their respective momenta, q_i their Cartesian coordinates, V the Coulomb interactions between the point charges, and E_j the electric field strengths in the x, y, and z directions. The other change that is required because of the inclusion of the electric field is to span the full two-dimensional space in the impact-parameter determination, rather than in just the $+y$ direction as done previously.[12]

The specific examples we chose to study were Rydberg atoms in the $n=10$ and $n=20$ states, and electric field strengths[13] of $\sim 10^9/(n+\Delta n)^4$ V/cm which classically field ionize hydrogenic Rydberg atoms in states $n'=n+\Delta n$, with $\Delta n=1$, 2, and 4. The collision velocity region spanned was $1 \leq v/v_e \leq 10$ where the orbital velocity of the Rydberg electron $v_e=n^{-1}$ a.u. or $2.19\times10^8 n^{-1}$ cm/sec, and v is the velocity of the incident ion relative to a stationary target atom. In this velocity region, the classical-trajectory Monte Carlo method has demonstrated it yields accurate cross sections for ion–hydrogenic-atom collisions. Also, classical studies of the field ionization properties of hydrogenic Rydberg atoms have been found to yield very accurate results,[13] but of course do not include quantum tunneling effects. Further checks on the excitation transfer cross sections were made with use of the analytical method of Lodge, Percival, and Richards.[14] To limit computer time we follow trajectories for a time corresponding to several electronic orbital revolutions. If at the end of this time the electron is in an n' state that would eventually be ionized, it is assumed to add to the ionization cross section.

The calculated results for collisions of protons with hydrogenic Rydberg atoms in constant electric fields for $n=10$ and $n=20$ states are displayed in Figs. 1 and 2. For all cases studied, the electron capture cross sections *decreased* with the application of an electric field. At the highest field strengths the electron-capture cross sections were found to decrease to approximately 25% of their zero-field values. Such collisional behavior can be understood because electron capture by a singly charged ion preferentially proceeds into a band of electronic levels which maximize around the n value of the reactant Rydberg atom.[6] We find electron capture followed by field ionization of the projectile plays the dominant role in the decrease of the cross section. This collisional behavior has been confirmed by following the time dependence of trajectories which lead to electron loss.

The ionization cross sections increased dramatically at all collision velocities. In the threshold region of low values of v/v_e, the ionization values rose to approximately an order of magnitude larger than the Rydberg-atom geometric value of $\pi n^4 a_0^2$ which equals 8.8×10^{-13} cm^2 for $n=10$ and 1.4×10^{-11} cm^2 for $n=20$. This increase was made up in part from the field ionization of the electron-capture component discussed in the previous paragraph. The most important contribution to the ionization process, however, arises from a two-step process consisting of excitation of the Ryd-

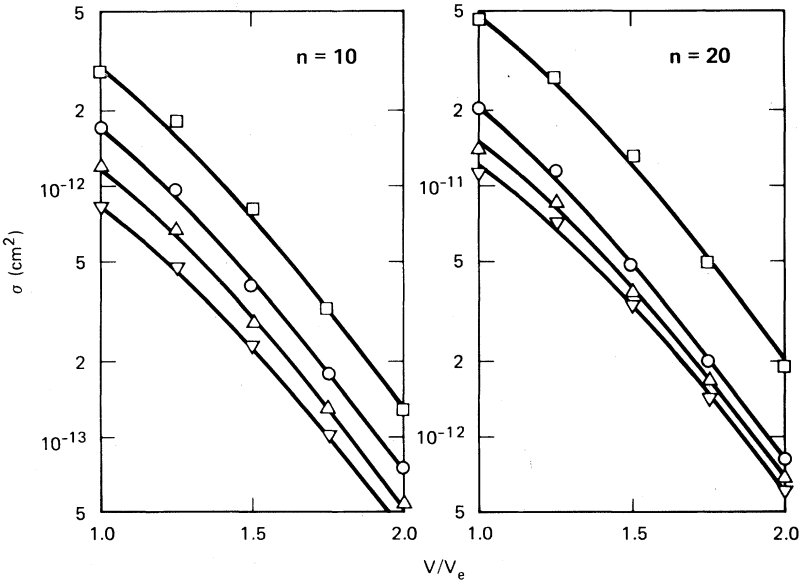

FIG. 1. Electron-capture cross sections for collisions of protons with hydrogenic Rydberg atoms in the $n=10$ and $n=20$ levels. The zero-field calculations are denoted by the squares, while the calculations performed with electric field strengths (Ref. 13) of $\sim 10^9/(n+\Delta n)^4$ V/cm, which classically ionize Rydberg atoms in the $n'=n+\Delta n$ ($\Delta n=1$, 2, and 4), are given by inverted triangles, triangles, and circles, respectively.

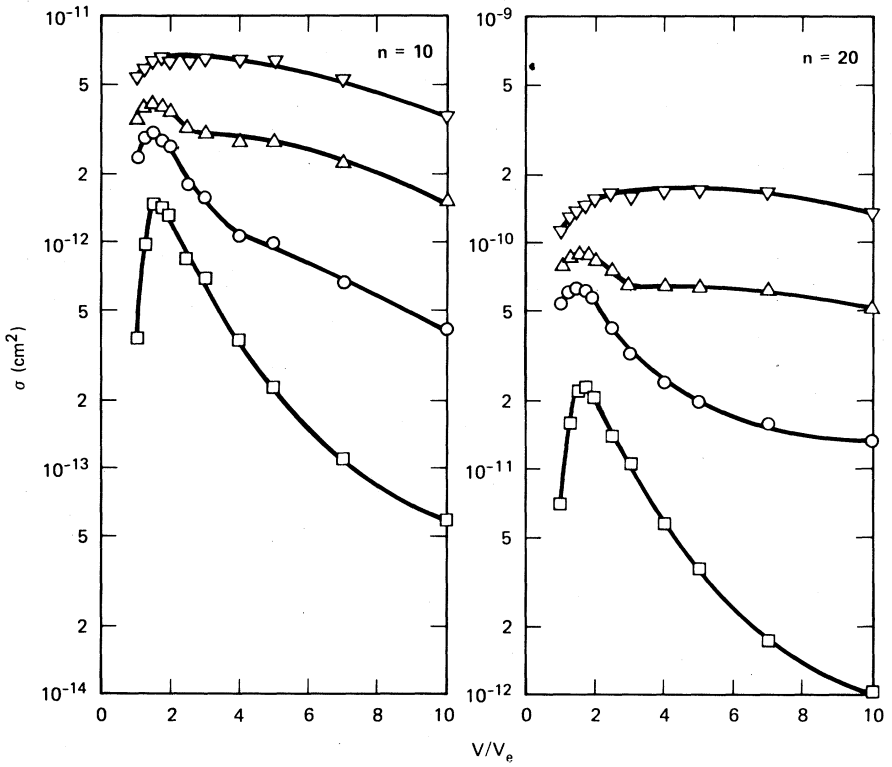

FIG. 2. Ionization cross sections for collisions of protons with hydrogenic Rydberg atoms in the $n = 10$ and $n = 20$ levels. The notation is the same as in Fig. 1.

berg atom to a higher level which is subsequently field ionized. Excitation cross sections, especially those for small changes in n, are larger than the Rydberg-atom geometric value at the collision velocities presented here. Thus, the ionization cross sections can be greatly enhanced by the application of an electric field. The largest percentage increases will be observed at values $v/v_e \lesssim 1.5$ where the zero-field ionization cross sections are decreasing rapidly with decreasing velocity and at $v/v_e \gtrsim 3.0$ where the zero-field ionization values are decreasing as v^{-2}.

We also investigated the dependence of the cross sections for various orientations of the electric field vector. Our coordinate system was defined such that the projectile traveled from the $-z$ to the $+z$ direction. As expected, the calculated cross sections were independent of the electric field strength orientation in the x-y plane. Small changes, however, were seen if the electric field was oriented along the same axis as that of the projectile, i.e., the $\pm z$ direction. The changes, though, were not outside the statistical errors in the calculation which were approximately $\pm 10\%$.

An interesting observation was made in the time dependence of collisions when the electric field was oriented in the $\pm z$ direction. If we computed the cross sections just after the collision encounter, but before the excited target or the highly excited electron-capture atoms were field ionized, significant differences were seen in the cross sections. When the field was set such that the target Rydberg electron was pulled toward the incoming projectile, both the ionization and electron-capture cross sections decreased relative to the zero-field case. However, if the field was set such that the electron was hidden behind the nucleus from the incoming projectile, both cross sections increased. We attribute this difference as due to whether the electric field decreased or increased the effect of the attractive Coulomb force felt on the target electron as the projectile retreated from the collision region into the $+z$ direction. An experimental observation of this behavior would require a pulsed electric field of such short duration that it appears to be infeasible at this time. Any longer pulse will yield cross sections similar to those presented because then there will be sufficient time to ionize the high-lying Rydberg atoms produced by the excitation

and electron-capture collision processes.

In conclusion, an initial attempt has been made to investigate the effects of strong dc electric fields on collisions of ions with Rydberg atoms. The changes in the calculated cross sections as a function of field strength are found to be quite large making experimental observations feasible. Hence, future studies of these processes should readily lead to progress in this subset of collision physics.

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Coherent Compton Effect

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Interference effects have been observed when coherently related x-ray beams are Compton scattered from an atomic system. We use dynamical diffraction methods to prepare the initial x-ray state and observe the coherent Compton effect in both cross-section and profile measurements.

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The conventional method of observing the Compton effect involves the preparation of an x-ray photon state of reasonably sharp momentum which is subsequently scattered from an electronic system. The final state of the total system consists of a recoiling electron and a scattered photon of reduced energy. If the target electron is initially at rest or in a state of uniform motion a unique relationship between photon scattering angle and energy loss results from simple kinematic considerations. However, if the electron is not free, but bound in an atom or solid, a range of photon energy losses will belong to a particular scattering angle. This range reflects the momentum spread in the bound-electron state. The total spectrum from all ground-state electrons in the target is commonly referred to as a Compton profile. Interest in this subject has continued into recent years precisely because electronic momentum distribution functions for atomic and solid-state systems can be extracted from Compton profiles.[1]

In the following we report on an extension of these notions to the case where the incident photon is no longer in a momentum eigenstate, but must be represented by a more complex coherent superposition of momentum eigenstates. From the general principle of superposition of quantum amplitudes we may anticipate the scattered photon intensity to be the sum of intensities expected from individual experiments for each momentum component in the incident state plus interference