Ionization of oriented elliptic Rydberg states by half-cycle pulses

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We calculate the ionization of elliptic Rydberg states by half-cycle unipolar electromagnetic-field pulses and examine the dependence of ionization probabilities on the eccentricity and the orientation of the ellipse with respect to the field direction of the pulse. It is suggested that the predicted large geometrical dependences be tested experimentally. [S1050-2947(97)02403-7]

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Recently sources of subpicosecond, unipolar ''half-cycle'' electromagnetic field pulses have been developed and employed to study the ionization of Rydberg atoms. These half-cycle pulses (HCP) were used first to study the ionization of Rydberg atoms with well-defined angular momentum quantum numbers [1]. More recently HCP have been used also to study the ionization of extreme parabolic states or Stark states [2]. Experiments and classical simulations by Jones *et al.* [2] for sodium atoms have shown marked difference between ionization probabilities of the two extreme Stark states within a given n manifold. A number of quantum calculations [3] have also confirmed that the ionization probabilities of the most redshifted (''downhill'') states are much larger than those for the most blueshifted (''uphill'') states. While these studies revealed a number of interesting features and experimental results appeared to have been confirmed by classical and quantum calculations, the full range of possibilities of the interaction of HCP with Rydberg atoms remained largely unexplored. For instance, the HCP field polarization has been directed only along the symmetry axis of the Stark states, thus the interaction is limited to cylindrical symmetry. In principle the polarization direction can be pointed along any direction with respect to the axis of the Stark states. Experimental data from such extended measurements would provide a more stringent test of our understanding of the interaction of HCP with Rydberg atoms. This is especially important for measurements when the precise HCP pulse shape is not well known.

In this paper we study the dependence of ionization probabilities of elliptic Rydberg states on the eccentricity and the orientation of the ellipse with respect to the polarization direction of the HCP. These elliptic states are examples of coherent states of a hydrogen atom [4] and can be prepared in the laboratory by laser excitation of Rydberg atoms in a combined mutually perpendicular weak electric and magnetic fields [5]. They are the most general stationary states of a hydrogen atom and include circular states and Stark states as special cases. Using a theoretical approach, we obtained a general expression from which the ionization probabilities for any elliptic states and any orientation of the HCP field polarization can be obtained *simultaneously*. Numerical results are presented for a number of field strengths for different orientations and eccentricities of the ellipse to stimulate future experiments. We point out that elliptic states have been created and used as targets for collisions with ions [6,7]. The dependence of electron capture cross sections of such elliptic states on the eccentricity and the orientation of the ellipse has been studied experimentally [6,7] and theoretically using either quantum calculations [8] or classical calculations [9,10].

Following Gay *et al.* [4], an elliptic state with eccentricity e within the n manifold of a hydrogen atom can be expanded in terms of the lm substates,

$$|n,e\rangle=\sum_{l,m} C_{nlm}(e)|nlm\rangle, \tag{1}$$

where the summation extends only over substates with even symmetry, i.e., $l+m$ is even. The explicit expression of C_{nlm} can be found in Ref. [4]. Such an elliptic state has an electronic density distribution that resembles that of a classical ellipse lying in the x-y plane with the x axis as the major axis. Note that the quantization axis of the spherical states is along the z axis. The special cases of $e=\pm 1$ are the extreme Stark states with the x axis being the symmetry axis. The circular state corresponds to the other special case of $e=0$.

We now consider the ionization of an elliptic state $|n,e\rangle$ by a fixed HCP taken to have the pulse shape

$$E(t)=F_s \sin^2(\pi t/T), \tag{2}$$

where T is the pulse duration. We used scaled field and scaled period, i.e., the field strength varies as n^{-4} and the period varies as n^3 for a Rydberg state with principal quantum number n. We explore the full range of parameters that can be varied, including the pulse shape, the period T, the principal quantum number n, and the eccentricity e and the orientation of the polarization of the HCP with respect to the ellipse. We will first fix the pulse shape to the form given in Eq. (2), and T to one scaled Kepler period and consider *all* orientations of the HCP and *all* eccentricities of the ellipse at the same time.

Consider a fixed xyz coordinate system where the classical ellipse is in the xy plane and the HCP polarization is along the z axis. Next we rotate the coordinate frames by the Euler angles $(\phi,\theta,0)$ such that the new polarization axis z'

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makes spherical angles (θ,ϕ) with respect to the fixed xyz frame. The transformation of spherical harmonics between the two frames is given by

$$Y_{lm}(\hat{\mathbf{r}})=\sum_{q} Y_{lq}(\widehat{\mathbf{r}'})D^{l}_{qm}(0°,-\theta,-\phi) \tag{3}$$

and the elliptic state is expressed in terms of spherical harmonics defined with respect to the new z' axis by

$$|n,e\rangle=\sum_{l,m}\sum_{q} C_{nlm}(e)D^{l}_{qm}(0°,-\theta,-\phi)Y_{lq}(\widehat{\mathbf{r}'})$$

$$=\sum_{l,q} d_{lq}(e,\theta,\phi)Y_{lq}(\widehat{\mathbf{r}'}) \tag{4}$$

where the last equation defines the coefficients d_{lq}. This equation describes an oriented ellipse. Since the time-dependent Schrödinger equation is linear, the ionization amplitude from an elliptic state, following Eq. (4), can be expressed as a linear superposition of ionization amplitudes from individual initial $|nlq\rangle$ substates. The dependence on the orientation angles are implicitly incorporated by the coefficients d_{lq} in the equation as well.

To calculate the ionization amplitude from an initial state $|nlq\rangle$, we solve the time-dependent Schrödinger equation of a hydrogen atom under the action of the HCP by direct numerical integration of the time-dependent equation using the split-operator-algorithm [11]. Fast Fourier transforms between position and momentum spaces are used to integrate over each time step. Since the HCP field is along the z' axis, the cylindrical symmetry of each $|nlq\rangle$ initial state allows us to do partial wave expansion of the time-dependent wave function

$$\psi(\mathbf{r},t)=\sum_{p=0}^{p_{\max}} R^{q}_{p}(r,t)Y_{pq}(\hat{\mathbf{r}}). \tag{5}$$

The convergence of expansion (5) is checked by the following: (i) the wave function does not hit the grid boundary in r during the time when the pulse is on; (ii) the norm of the highest angular momentum partial wave is negligible. We found $p_{\max}=40$ is adequate for scaled fields up to $F_s=0.5$. By projecting Eq. (5) for $t\geqslant T$ into continuum Coulomb wave functions $|\epsilon,pq\rangle$, the ionization amplitude to a final continuum state $|\epsilon,pq\rangle$ from an initial Rydberg state $|nlq\rangle$ is obtained. These amplitudes are combined with the d coefficients in Eq. (4) to obtain ionization probability amplitude to a continuum state $|\epsilon,pq\rangle$ from an initial elliptic state with eccentricity e and orientation (θ,ϕ). The total ionization probability is obtained after integrating over the continuum electron energy and summing over the partial waves.

The procedure outlined in the previous paragraph requires that the ionization *amplitudes* from all the initial $|nlq\rangle$ $(q\geqslant 0)$ substates be calculated, which would then give ionization probabilities for *any* orientations of the HCP field polarization as well as *any* eccentricities of the ellipse. To perform calculations for actual Rydberg states where experiments are likely to be done, the amount of numerical calculations will be enormous. Fortunately the HCP ionization probabilities are relatively insensitive to the principal quantum number n of the initial state. As shown by Reinhold and Burgdörfer [3] for the HCP ionization of Stark states, the two calculations for $n=5$ and $n=10$ agree reasonably well for the same scaled peak field and pulse width, showing the weak n dependence. Similar calculations [8] for electron capture from $n=4$ and 5 states have been carried out for proton-hydrogen collisions and the results were shown to have similar geometric dependence for experiments carried out for $Na^{+}+Li(n=25,e)$ collisions if the collision velocity is scaled. With this in mind, we anticipate that the results presented below, which were carried out for initial $n=4$ states, are valid in describing the HCP ionization of higher n elliptic states, so long the same scaled field strength and pulse shape are used.

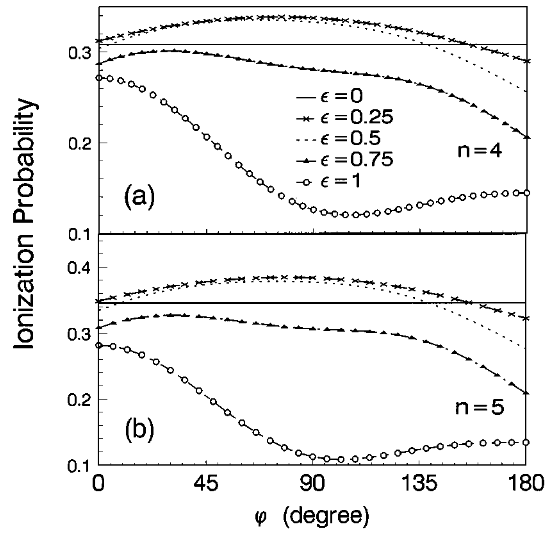

FIG. 1. Ionization probabilities vs ϕ for $\theta=90°$ from (a) $n=4$ and (b) $n=5$ elliptic states by HCP pulses. The scaled peak field strength is $F_s=0.3$ a.u.

As an illustration of the weak n dependence of the ionization probabilities, in Fig. 1 we compare the ϕ dependence for $n=4$ and $n=5$ elliptic states for the scaled field strength of $F_s=0.3$, with fixed $\theta=90°$, for the eccentricities indicated. They correspond to configurations where the HCP field polarization is lying in the plane of the ellipse but is rotated from parallel to the axis continuously to the antiparallel direction. It is quite evident that the ϕ dependence and the ionization probabilities in the two frames are nearly the same, verifying the weak n dependence of the initial state. Thus in the following only results for $n=4$ states will be shown. These figures show that the ϕ dependence is stronger for states with eccentricities larger than 0.5. We mention that existing theories [3] and experiment [2] have examined only the two special cases in this figure, namely, $e=1$, $\phi=0°$ and $e=1$, $\phi=180°$. The former corresponds to a downhill Stark state and the latter an uphill Stark state if we view the field direction as being fixed. We also note that for circular states $(e=0)$ there is no ϕ dependence as expected from the symmetry.

We next examine the orientation and eccentricity dependence of the ionization probabilities as the scaled field strength is varied. While in general the ionization probabilities increase with increasing scaled field strengths, the ϕ dependence does vary significantly. In Fig. 2 we show the

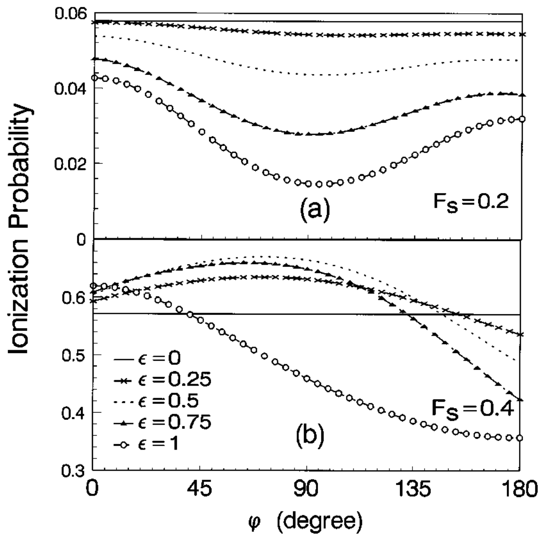

FIG. 2. Ionization probabilities vs ϕ for $\theta=90°$ from $n=4$ elliptic states for $e=0.0$, 0.25, 0.5, 0.75, and 1.0 for scaled peaked field strengths of $F_s=0.2$ and 0.4 a.u.

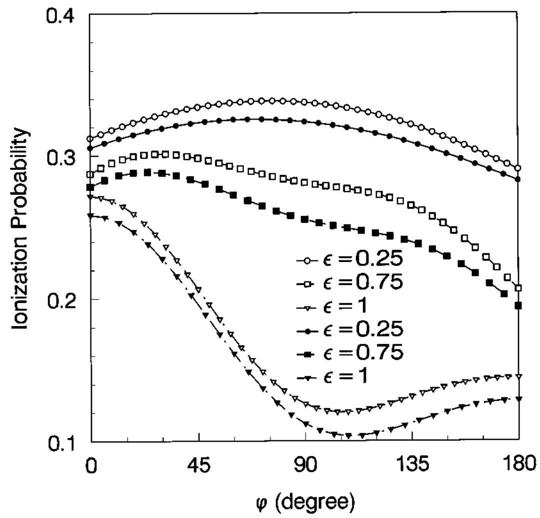

FIG. 4. Dependence of ionization probabilities vs ϕ with the HCP pulse shapes, for $\theta=90°$ from the $n=4$ elliptic states for $F_s=0.3$ a.u. Open symbols, from Eq. (2); solid symbols, pulse shape from Ref. [12].

results for $F_s=0.2$ and 0.4. For $F_s=0.2$, the ionization probability tends to have a minimum for $\phi=90°$ and is nearly symmetric. This is also true for $F_s=0.1$ (not shown). At the higher fields [$F_s=0.3$, see Fig. 1(a)], ionization is larger when the field direction is on the downhill side where the electronic charge density is higher. This change of ionization probabilities with the eccentricity and the scaled field strength is rather rapid and thus provides a more sensitive test of the models or calculations on HCP-atom interactions than have been studied thus far.

Our formulation allows us to explore ionization probabilities for any orientation of the HCP polarization field direction. As an example, consider $\phi=0°$ and vary θ from 90° to $-90°$, see Fig. 3. This corresponds to the electric field direction being rotated around the minor axis of the ellipse, from the direction of the major axis out of the plane until it is antiparallel to the major axis. For positive θ the HCP field is pointing toward the downhill side and for negative θ the field is pointing toward the uphill side. The θ dependence does change significantly with the scaled field strength, and with the eccentricity, especially for negative θ. Note that the probabilities are symmetric with respect to $\theta=0°$ for circular states, as expected from the geometry. For $e=1$, the $\theta=90°$ and $-90°$ correspond to situations studied by experimentalists and theorists thus far. It is clear that only a very small range of the parameter space has been explored until now.

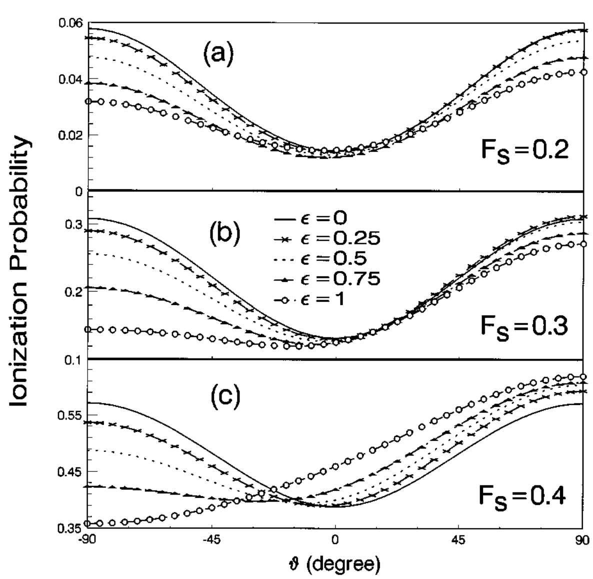

FIG. 3. Ionization probabilities vs θ (for $\phi=0°$) and eccentricity e for $n=4$ elliptic states. The scaled peak field strengths are $F_s=0.2$, 0.3, and 0.4 a.u.

We have illustrated the θ and ϕ dependence of ionization probabilities for different eccentricities. Depending on the experimental setup, it is possible to study the eccentricity dependence for a fixed HCP field direction as well. This may be desirable since only the magnitude of the electric or magnetic fields for creating the elliptic states are modified without changing their directions. Actual comparison with experiment is not possible until experimental parameters are known. However, by suitably exploring the full range of parameters offered in the present formulation (the θ, ϕ, and e dependences), a detailed test of the accuracy of the calculations and consistency of experiments would become possible.

We have also examined whether the calculated results are sensitive to the precise HCP pulse shape by carrying out calculations using the pulse shape of Bugacov *et al.* [12], which was to simulate the experimental pulse shape of Jones *et al.* [1,2]. The resulting ϕ dependence of the ionization probabilities for $\theta=90°$ for three different eccentricities are shown in Fig. 4 for $F_s=0.3$. Clearly the pulse shape dependence is rather weak. This is also true for the θ dependence for a fixed ϕ (not shown). In general the orientation and the eccentricity dependence for the same scaled field strength but somewhat different pulse shape is essentially the same. We mention that for the circular state ($e=0$) the ϕ dependence in Fig. 4 is a constant. The calculated ionization probability for $e=0$, using the pulse shape of Bugacov *et al.* [12] is

0.302 while using the pulse shape of Eq. (2) it is 0.308.

In summary, we have shown how the ionization probabilities of elliptic Rydberg states in general depend on the eccentricity and the orientation angle of the ellipse with respect to the direction of polarization of the half-cycle electromagnetic pulses. Although the calculations have been carried out for $n=4$ elliptic states, we have also illustrated the weak n dependence and predicted that these dependences are also valid for higher elliptic Rydberg states where experiments can be carried out. The predicted geometric dependence is quite large and can be easily tested in experiments. We have also shown that the geometric dependence examined here is relatively insensitive to the HCP pulse shapes used and the calculated results can be used as a guide for designing experiments where the geometric dependence is most drastic. Finally it is desirable to test the results presented here using classical theories which have been used successfully for studying HCP ionization for high Rydberg states [1,2].

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