Excitation in Ion-Atom Collisions Inside Subfemtosecond Laser Pulses

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We discuss new excitation mechanisms in energetic ion-atom collisions embedded in short laser pulses. For comparable duration and strength of the pulse and collisional interaction, the laser field will probe and modify the interaction between projectile and target. Coherence effects emerge, insight into reaction dynamics is gained, and new dynamical features are discovered. As an example, we show (i) how a propensity rule for *s-p* excitation can be dramatically changed, and (ii) how the presence of the laser pulse modifies the ionization process in ion-atom collisions.

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Ion-atom collisions, and atoms in strong laser fields, have been studied separately for many years, and research in these areas has contributed significantly to our present understanding of atomic dynamics. Powerful momentum space imaging techniques for kinematical complete studies have paved the way for extremely detailed investigations of ion-atom collisions [1], light sources with high intensity and pulse durations in the femtosecond range are now available [2], and, in strong field physics, modeling and *ab initio* calculations have joined forces for theoretical progress [3]. In this Letter, we explore novel effects from energetic ion-atom (at keV projectile energies) collisions taking place in a pulsed laser field of duration comparable to the collision time (of the order of fs). Particularly interesting from the theoretical point of view is the possible interference of the two interactions.

Two new developments indicate that phenomena analogous to those in the present investigation might soon become subjects of experimental work. First, atomic excitation induced by a subfemtosecond soft x-ray pulse in the presence of a fs laser pulse has been reported [4]. Second, precision recoil experiments with cold targets make it possible to detect previously hidden details of atomic collisions from the extremely accurate determination of the recoil momenta (see, e.g., [1,5–7]).

Of special interest for this work are the recoil-ion momentum measurements on single and double ionization of atoms by intense laser pulses [6,7]. These experiments successfully reveal the mechanisms for enhanced double ionization [8] even though, for kinematical completeness, they must be done with at most one atom in the laser focus per shot. Adding to these experiments, collision interactions by ions or electrons, in order to bring out the new effects reported here, require a high density pulsed projectile beam. Such beams are presently available, and in the future the laser repetition rate will increase. Note that theoretical work on laser-assisted $(e, 2e)$ [9], and laser-assisted charge-transfer processes [10] has been reported.

At the energies considered here, excitation and ionization mechanisms in ion-atom collisions are dipole dominated and have large probabilities [11], and the electronic problem for the collision embedded in a short laser pulse is well described by the time-dependent Schrödinger equation $i\partial_t\Psi(\mathbf{r}, t) = H(t)\Psi(\mathbf{r}, t)$ within the impact parameter and dipole approximations [atomic units (a.u.) are used throughout unless otherwise indicated]. Here $H(t) = h(\mathbf{r}) + V_p(t)$, with $h(\mathbf{r})$ the effective one-electron Hamiltonian, and $V_p(t)$ a superposition,

$$V_p(t) = -\frac{Z_p}{|\mathbf{R}(t) - \mathbf{r}|} - \mathbf{E}(t)\cdot\mathbf{r}, \tag{1}$$

of contributions from the passing ion following the trajectory $\mathbf{R}(t)$, and the laser field $\mathbf{E}(t) = f(t)E_0\boldsymbol{\epsilon}\cos(\omega t - \delta)$, with E_0 the field strength, $f(t)$ the pulse shape, $\boldsymbol{\epsilon}$ the polarization vector, ω the laser frequency, and δ the phase of the field. In the calculations, we use a finite $\sin^2$-pulse, $f(t) = \sin^2\pi(t/\tau - 1/2)$ for $-\tau/2 \leq t \leq \tau/2$ and 0 otherwise. Note that τ and/or δ are adjusted to secure a zero dc field component of the propagating pulse [12].

After a multipole expansion of $Z_p/|\mathbf{R}(t) - \mathbf{r}|$, the interaction reads $V_p(t) \approx -\mathbf{r}\cdot[\mathbf{E}(t) + \mathbf{E}_c(t)]$, where $\mathbf{E}_c(t) = Z_p\mathbf{e}_{\mathbf{R}(t)}/R^2(t)$. Interference effects are expected to be most pronounced when the two dipoles are of equal magnitude. For $Z_p = 1$ and for a typical impact parameter, $b \sim 5$, this corresponds to laser intensities $\sim10^{14}$–10^{15} W/cm^2.

Here we first explore interference effects by considering direct *s-p* excitation of the target atom. In the combined process of Fig. 1, the collision plane is defined by the impact parameter $\mathbf{b} = b\mathbf{e}_y$, and the trajectory $\mathbf{R}(t) = \mathbf{b} + \mathbf{v}t$. A linearly polarized laser pulse with polarization vector parallel to the impact parameter, $\boldsymbol{\epsilon} = b\mathbf{e}_y$, is propagating along the quantization axis (z) perpendicular to the collision plane. This situation sets up a collision induced time-dependent electric field in the xy plane as well as a laser field in the y direction. The laser field and the dipole part of the collision field in the y direction are shown in Fig. 1 (left). The fields are of similar magnitude. The laser field contains one cycle in the present example.

Coupling among excited p states can be safely neglected, so the dynamics is determined by

$$\begin{pmatrix} i\partial_t c_s \\ i\partial_t c_{p-} \\ i\partial_t c_{p+} \end{pmatrix} = \left[f_{sp}(R) \begin{pmatrix} 0 & \text{c.c.} & \text{c.c.} \\ e^{-i[\Delta E_{sp}(t)-\phi(t)]} & 0 & 0 \\ e^{-i[\Delta E_{sp}(t)+\phi(t)]} & 0 & 0 \end{pmatrix} + y_{sp}E_0 f(t) \begin{pmatrix} 0 & \text{c.c.} & \text{c.c.} \\ -e^{i\Delta\varepsilon_{sp}t}\cos(\omega t+\delta) & 0 & 0 \\ e^{i\Delta\varepsilon_{sp}t}\cos(\omega t+\delta) & 0 & 0 \end{pmatrix} \right] \begin{pmatrix} c_s \\ c_{p-} \\ c_{p+} \end{pmatrix}, \quad (2)$$

where c_s, c_{p-}, c_{p+} are amplitudes for the s, $p_{m=-1}$, and $p_{m=+1}$ states, respectively. The matrix elements of the collision part are factorized into a part which depends only on the internuclear distance $f_{sp}(R)$ and a part which depends on two time-dependent phases. One phase, $\phi(t)$, varies from $-\pi$ to 0 during the collision. The other, ΔE_{sp}, is given by the asymptotic energy difference between s and p states, $\varepsilon_p - \varepsilon_s$, and the distortion of each state, $\Delta E_{sp} = \int_{-\infty}^{t}(\langle p|V(t')|p\rangle - \langle s|V(t')|s\rangle)dt' + (\varepsilon_p - \varepsilon_s)t \approx (\varepsilon_p - \varepsilon_s)t = \Delta\varepsilon_{sp}t$. The second matrix describes laser induced couplings, and y_{sp} is the dipole matrix element $\langle s|y|p\rangle$ between s and p states.

In collision physics, a propensity rule for $p_\pm$ excitation is easily explained by noting that the p_+ rotates anticlockwise around the z axis in Fig. 1 while the p_- state rotates clockwise. Velocity matching shows that p_- (p_+) excitation should dominate for positive (negative) b. The rule was originally derived by observing that a stationary phase nearly occurs for the c_{p-} amplitude when $\Delta\epsilon_{sp}t - \phi(t) \approx 0$ [13]. As may be seen from Fig. 1, and the range of $\phi(t)$, the maximum transition probability to the p_- state occurs when the collision range a and velocity v are connected by

$$\Delta\epsilon_{sp}a/v \sim \pi. \quad (3)$$

Here a is typically a few a_0 and typically smaller than $\sim 10a_0$ (see also the range of impact parameter with efficient excitation probability in Figs. 2–4). In the parameter range determined by (3), the population of the p_+ state will, on the other hand, be suppressed. These stationary phase predictions were confirmed by nonperturbative calculations [14].

The effect of the laser when added to the collision dynamics may be explored analytically by assuming ω to be nearly resonant with the s-p transition, such that the rotating wave approximation applies. During a certain period, the collision and laser phases are approximately stationary, and the first-order amplitude for coherent s-p_- excitation reads

$$c_{p-}(t) \approx f_{sp}(R) - y_{sp}E_0 f(t)e^{i\delta}. \quad (4)$$

If the two terms in (4) are approximately equal in magnitude and the phase $\delta = 0$, the probability for exciting the p_- state will vanish in the region where the collision process alone favors a strong excitation. The collisionally suppressed c_{p+} amplitude is in the same region given by

$$c_{p+}(t) = \int_{-\infty}^{t} [f_{sp}(R)e^{-i[\Delta E_{sp}(t')+\phi(t')]} + y_{sp}E_0 f(t')e^{i\delta}]dt', \quad (5)$$

which may lead to more excitation than induced by the laser alone.

In order to explore these predictions, we carry out two sets of calculations. First, we consider a four state minimal-model calculation for a proton colliding with a hydrogen target including the $1s$ and $n = 2$ levels. Figure 2 shows the excitation probability for H($2p_-$) with and without a laser pulse with $\tau = 0.3$ fs and peak intensity such that

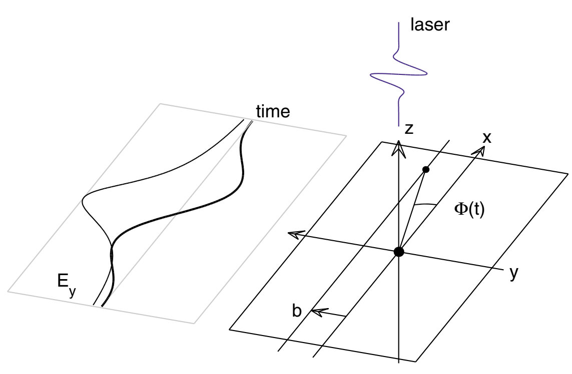

FIG. 1 (color online). Right: The collision plane (xy) is defined by the impact parameter $\mathbf{b} = b\mathbf{e}_y$ and the projectile trajectory $\mathbf{R}(t)$. The angle of the projectile $\Phi(t)$ defines its position. The laser is propagating perpendicular to the scattering plane and parallel to the quantization axis z. Left: The laser is linearly polarized along the y direction. The y components of the dipole part of the collisionally induced field (thin line) and the laser field (thick line) are shown.

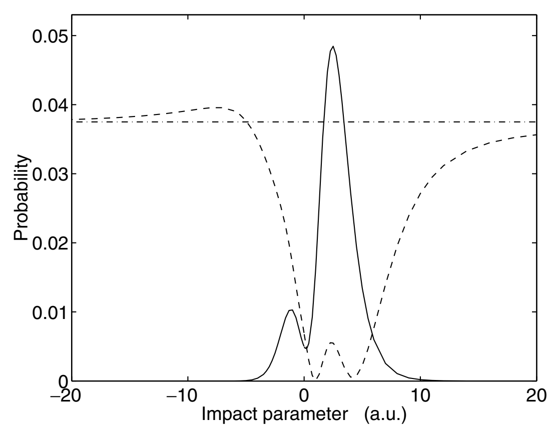

FIG. 2. Excitation probability for H($1s$)-H($2p_-$) ($Z_p = 1$) in the presence (dashed line) and absence (full line) of a laser pulse. The dot-dashed line is the laser-only contribution. The projectile velocity is $v = 1$ a.u., the duration of the laser pulse is $\tau = 0.3$ fs, and the peak intensity is set by $y_{sp}E_0 = 0.045$ a.u. (cf. Fig. 4).

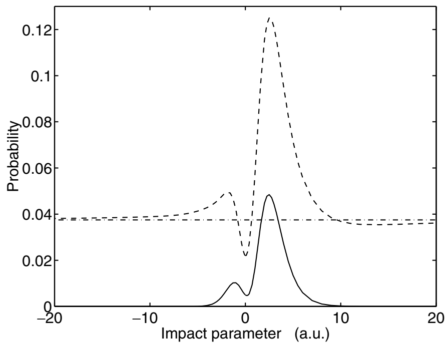

FIG. 3. As Fig. 2, but for constructive interference between the collision and laser interactions (see text).

$y_{sp}E_0 = 0.045$ a.u. The projectile velocity is 1 a.u. which, from the propensity rule (3), is close to the value for maximum excitation. For a collision-only process, we observe a strong propensity for excitation at positive b [left-hand side (lhs) of the target as seen from the approaching projectile] as compared to the p_+ state, which is equal to the p_- excitation probability at the right-hand side. The laser-excitation probability in absence of an ion is shown by the horizontal line. Note that for the present field strength ionizing two-photon absorption can be neglected (from nonperturbative grid calculations, we have estimated this probability to be less than 0.001). In the combination of both laser and collision, the situation is dramatically changed. For positive b, precisely where the s-p_- process is favored without a laser field, the excitation probability vanishes. For negative b (corresponding to s-p_+ excitation at positive b), the excitation probability becomes larger than the contribution from the laser alone. These findings are in complete agreement with the above analysis: Left side collision-only induces a negative angular momentum with respect to the z axis, while a combined event leads to a preferred positive angular momentum of the target atom. A surprising feature is observed in Fig. 2: a strong modification of the laser-excitation probability at large b (the corresponding collision-only process has zero probability). This effect results from a coherent interplay between a weak collisional long range interaction on top of the much stronger interaction between target and laser field. Thus, a probe of long range interaction of atomic collisions can be read out of the laser-excitation probability.

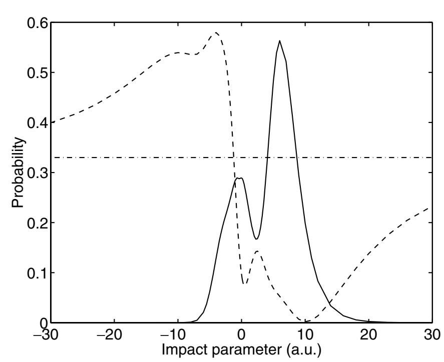

FIG. 4. As Fig. 2, but for Na(3s)-Na(3p_-), $\tau = 1$ fs, and $v = 0.4$ a.u. The peak intensity of the laser is set by $y_{sp}E_0 = 0.1$ a.u., corresponding to the peak value in the collisional strength $f_{sp}(R)$.

In Fig. 3, we show the corresponding results obtained by switching the phase δ by π. Because of constructive interference, cf. Eq. (4), the orientation asymmetry is dramatically increased. The maximum p_- probability is roughly twice the incoherent sum of the individual contributions.

In the second set of calculations, we consider s-p excitation in Na(3s) [15], and Fig. 4 shows the propensity rule for s-p_- excitation. The inversion of the rule is even more pronounced: At positive b, the excitation probability nearly vanish in the range where the field-free excitation is strong. At negative b, the laser-excitation probability is enhanced by almost a factor of 2. The long range effects are also much more dramatic. The laser-excitation probability is still significantly changed at impact parameters where a collision-only process would not excite the target atom.

Detection of these processes can, in principle, be done when the laser field is slightly detuned from the s-p energy difference. A triple coincidence between the decaying atom, the projectile, and the recoil of the target atom would display the details of the collision process as outlined above. Coincidence experiments involving neutral atoms are, however, extremely difficult. Therefore the effects reported here might be most readily detected in ionization processes leading to three charged fragments. Depending on the relative phase between the interactions, the charged fragments can be expected to have different momenta, and the details of the collision process including relative phases may therefore be traced in advanced recoil momentum spectroscopy [5]. For the fields and collision energies considered here, a collision will lead to much more energetic electrons than breakup induced by a short laser pulse. We suggest that the direction of the electron and proton momenta in coincidence can be traced back to a unique phase of the actual collision.

To quantify these ideas, we have performed classical trajectory Monte Carlo (CTMC) simulations of the ionized electron momenta. The laser pulse is similar to the one in Fig. 1, but 4 times more intense to obtain an ionization probability comparable to collisional ionization. Figure 5 shows results of simulations with a microcanonical selection of initial electron positions and velocities combined with a uniform selection of positive in-plane impact parameters. The upper part of Fig. 5 shows xy plane electron momenta for the laser-only case. The laser field causes an asymmetric scattering along the direction of the polarization vector y. This asymmetry is unique for ultrashort pulses, and it reflects a relation between the direction of ejection of the electron and the initial phase of the field [16].

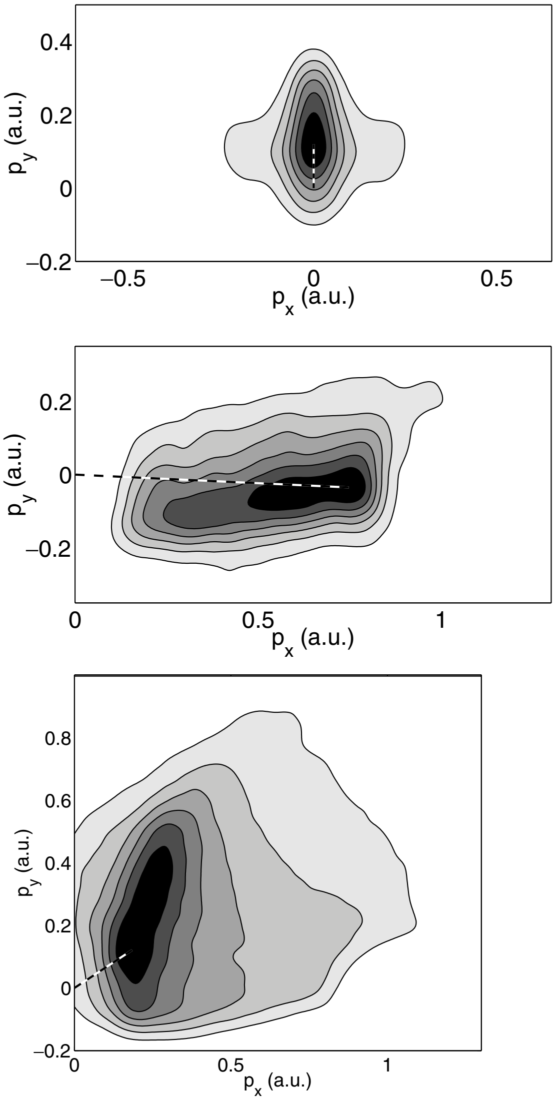

FIG. 5. Distribution of the ejected electron momenta in the collision plane for ionization in p-H($1s$). Upper: Laser only. Middle: Collision only. Lower: Collision and laser. The CTMC data have been binned into a 32×32 array and slightly smoothed. Each new shade corresponds to an increase in probability density by 15%. The broken lines indicate the position of the most probable momentum. Parameters as in Fig. 2, except $E_0 = 0.19$ a.u.

The middle panel of Fig. 5 shows the collision-only case. The electron momenta have a range of positive x components corresponding to electrons ionized in the beam direction. The forward scattering of ionized electrons including saddle point mechanisms are well known experimentally [17] and theoretically [18]. The lower panel of Fig. 5 shows a situation in combined fields for a particular choice of phase δ. A completely different momentum spectrum is observed with a localization towards positive x and y components. This corresponds to electron emission in the beam direction on the lhs of the target, thus completely spatially separated from the collision-only and laser-only processes. We have performed a series of simulations in combined fields for different values of δ. For each choice, one obtains a unique momentum distribution. Changing the phase by π, e.g., gives a distribution centered in the negative plane.

By simply detecting coincidences between the momenta of the charged fragments, details on subfemtosecond processes can be exposed. The detailed analysis based on quantum mechanical calculations of the electron emission recoil characteristics will be explored in the near future. Our results show that one-electron dynamics in two external fields may lead to new and interesting coherence effects to the same extent as observed in correlated two-electron dynamics in a single strong field [5–7].

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