Manipulating Ion-Atom Collisions with Coherent Electromagnetic Radiation

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Laser-assisted ion-atom collisions are considered in terms of a nonperturbative quantum mechanical description of the electronic motion. It is shown for the system He^{2+}-H at 2 keV/amu that the collision dynamics depend strongly on the initial phase of the laser field and the applied wavelength. Whereas electronic transitions are caused by the concurrent action of the field and the projectile ion at relatively low frequencies, they can be separated into modified collisional capture and field ionization events in the region above the one-photon ionization threshold.

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Thanks to significant experimental and theoretical progress in recent years, the understanding of basic atomic collision processes has reached a state of high maturity. Experimental techniques, such as the combined electron and recoil-ion momentum spectroscopy made it possible to study the details of electron, photon, and ion collisions with atomic and molecular targets, and to identify the operation of different scattering mechanisms [1].

Refined theoretical methods to calculate differential and integrated cross sections have contributed decisively to the interpretation of the data. Naturally, theory has been most successful in the description of three-body problems, such as electron-induced [2] and photon-induced [3] ionization of hydrogen atoms, or excitation, ionization, and electron capture in (effective) one-electron ion-atom scattering systems (see, e.g., Ref. [4] and references therein). These processes can be calculated with high accuracy by modern algorithms for the solution of the Schrödinger (Dirac) equation that governs the motion of the particles.

Given these powerful experimental and theoretical techniques, it seems timely to reconsider a somewhat more complex problem that was first addressed more than 20 years ago on the basis of rather restrictive theoretical models: the modification of electronic processes in ion-atom collisions in the presence of coherent electromagnetic radiation. Such laser-assisted collisions are interesting for several reasons: First, they might exhibit new structures in the observable quantities, such as final charge-state distributions and electron spectra, and might broaden our general understanding of dynamic atomic processes. Second, it might be possible to control the interaction pathways in ion-atom collisions, i.e., to enhance the population of favored final states and to suppress the production of undesired ones by a suitable choice of the laser parameters. This could be useful for a variety of applications, such as laser-driven fusion or plasma heating.

In fact, little is known about the physics and potentialities of laser-assisted ion-atom collisions to the present date. This is due to the restrictive nature of the previous theoretical models and the absence of experimental investigations apart from studies with laser-excited targets (see, e.g., Ref. [5] and references therein), and a few considerations of laser-assisted capture at thermal collision energies [6]. The early theoretical works mentioned above focused on field-assisted capture in slow collisions and were based on very limited expansions of the electronic wave function in terms of atomic [7] or molecular (e.g., Ref. [8]) states. The coupling to the continuum, which should be important except for rather weak fields was not taken into account. Recently, first-order perturbation theory was used to study electron capture [9] and ionization [10] in fast collisions assisted by relatively weak fields of low frequency.

In the present contribution, the problem at hand is investigated on the basis of a more elaborate theoretical approach that allows the *simultaneous* description of capture and ionization processes. It is the purpose of this work to elucidate the subtle dynamics of laser-assisted ion-atom collisions in closer detail and to guide future experimental activities, which seem feasible with present-day technology. To this end, the nonperturbative basis generator method (BGM) [11,12], which has been very successful for the description of field-free ion-atom collisions (for a review of applications, see Ref. [13]), is adapted to study the He^{2+}-H system in the presence of a laser field.

Let us start from the semiclassical approximation; i.e., the laser interaction and the Coulomb field of the projectile ion are taken into account in terms of classical, time-dependent potentials. In the length gauge, the Hamiltonian for a one-electron system reads (in atomic units, $\hbar = m_e = e = 1$)

$$\hat{H}(t) = -\frac{1}{2}\Delta - \frac{Q_T}{r} - \frac{Q_P}{|\mathbf{r} - \mathbf{R}(t)|} - \mathbf{r}\cdot\mathbf{E}(t), \quad (1)$$

where Q_T and Q_P are the charges of the target and projectile nuclei, respectively. The rectilinear trajectory of the projectile ion $\mathbf{R}(t)$ is characterized by the impact-parameter b and the constant velocity v_P via $\mathbf{R}(t) = (b, 0, v_p t)$. The electric field $\mathbf{E}(t)$ polarized along the direction $\boldsymbol{\epsilon}_{\rm pol}$ is given in the dipole approximation by

$$\mathbf{E}(t) = \boldsymbol{\epsilon}_{\rm pol} E_0(t) \sin(\omega t + \delta), \quad (2)$$

where ω and δ are the frequency and the initial phase. In

this work, only square pulses, i.e., $E_0(t) = E_0$, are considered. The time-dependent Schrödinger equation (TDSE) $i\partial_t|\Psi(t)\rangle = \hat{H}(t)|\Psi(t)\rangle$ for the Hamiltonian (1) is solved with the BGM, which aims at a finite representation of the state vector $|\Psi(t)\rangle$ in terms of a dynamically adapted basis. In the present work, the basis includes all undisturbed target eigenstates $|\varphi_v^0\rangle$ of the *KLMN* shells, and pseudostates, which are constructed by repeated application of the (regularized) projectile potential W_P onto the set $\{|\varphi_v^0\rangle\}$:

$$|\chi_v^\mu(t)\rangle = [W_P(t)]^\mu|\varphi_v^0\rangle \qquad \mu = 1, \ldots, M = 8. \quad (3)$$

The pseudostates are orthogonalized to the undisturbed functions such that their population at asymptotic times after the collision can be interpreted as total electron loss P_{loss} from the target. The capture contribution P_{cap} is extracted by projection onto the moving projectile states of the *KLM* shells, and the total ionization probability P_{ion} is calculated as $P_{\text{ion}} = P_{\text{loss}} - P_{\text{cap}}$. The basis (3) is well founded for field-free ion-atom collisions [12], and, in particular, it was applied successfully to the He^{2+}-H system [14]. For laser-assisted collisions, the coupling to the complementary part of the Hilbert space is minimized with this construction in the same sense as in the field-free case. This is easily shown and will be detailed in a forthcoming paper.

Notwithstanding this formal justification, it is appropriate to check explicitly whether the basis (3) is capable of describing interactions with a laser field. To this end, some test calculations were performed for a hydrogen atom exposed to an electromagnetic field *without* additional ion impact. The TDSE was propagated over 300 a.u. of time, and the time-resolved results for the depopulation of the ground state and for target excitation were compared with calculations of Ref. [15], in which the state vector $|\Psi(t)\rangle$ was expanded in angular momentum components. Good agreement was found for the frequency $\omega = 0.2$ a.u. and the field strengths $E_0 = 0.01$ and 0.1 a.u.

Let us now consider laser-assisted He^{2+}-H collisions at the projectile energy $E_P = 2$ keV/amu ($v_P = 0.283$ a.u.), where capture is the dominant process in the field-free case [16]. We keep the wavelength of the example above ($\omega = 0.2$ a.u. corresponds to $\lambda = 228$ nm), choose the field intensity $I = 1.4 \times 10^{13}$ W/cm^2 (i.e., $E_0 = 0.02$ a.u.), and longitudinal polarization $\boldsymbol{\epsilon}_{\text{pol}} \| v_P$. The TDSE is propagated over 300 a.u. of time corresponding to the initial and final separations $R_{i,f} = 45$ a.u. between the projectile and target nuclei. These parameters ensure that field ionization in the absence of the projectile ion is negligible [17].

Figure 1 shows impact-parameter-weighted capture probabilities for the initial phases $\delta = 0$ and $\delta = \pi$, as well as the field-free and the phase-averaged results. The latter are obtained from calculations for $\delta = 0, \pi/2, \pi, 3\pi/2$, and are included to study the net effect of the laser field, since it seems impossible to control δ experimentally. The results for laser-assisted capture differ significantly from the field-free case such that the averaged

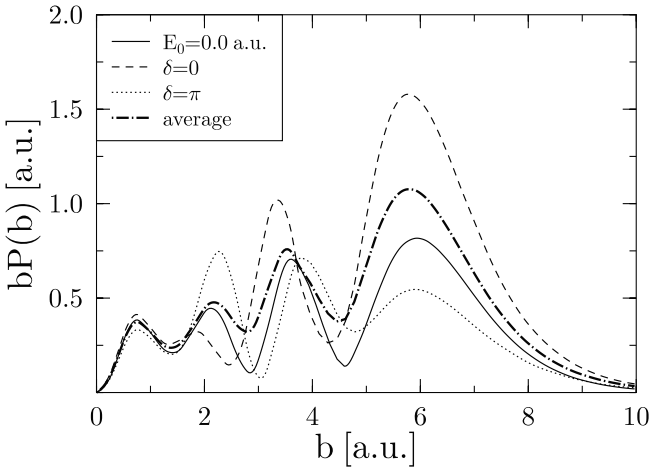

FIG. 1. Impact-parameter-weighted total capture probabilities as functions of the impact parameter b for He^{2+}-H collisions at $E_P = 2$ keV/amu, laser field strength $E_0 = 0.02$ a.u., frequency $\omega = 0.2$ a.u., and longitudinal polarization $\boldsymbol{\epsilon}_{\text{pol}} \| v_P$. Results for different initial phases δ and the phase average are displayed.

probability is enhanced except for small impact parameters. The total field-assisted capture cross section is 27% larger than the field-free one.

The results depend strongly on the initial phase δ. The most significant variations are observed for the two phases displayed in Fig. 1. They occur at relatively large impact parameters b and can be related to the expectation value $\langle z(t)\rangle = \langle\Psi(t)|z|\Psi(t)\rangle$ of the electronic coordinate in the longitudinal direction. The BGM calculations for $\langle z(t)\rangle$ show that the bound electron simply oscillates in the electromagnetic field without ion impact. In the other limiting case, the field-free collision, the electron cloud is pulled toward the projectile in the ingoing channel before a considerable fraction is transferred around the closest approach ($t = 0$). When the ion collision and the laser interaction are combined, the efficiency of capture depends crucially on the interplay between both movements: In the case of $\delta = 0$, the field-induced oscillation and the ion-induced polarization are synchronized around the closest approach; i.e., both fields have a component that pulls the electron cloud in the $-z$ direction for $t < 0$ and in the forward direction for $t > 0$. As a consequence, capture is enhanced. For $\delta = \pi$ capture is reduced, since laser and projectile fields counterbalance each other.

These arguments apply only at relatively large impact parameters. At small to intermediate b, the dynamics are more involved as capture is associated with electronic oscillations between the nuclei, and a synchronization with the field oscillations cannot be achieved easily. Therefore, the phase dependence of the capture probability does not show a uniform pattern in this region.

The observed (anti-)synchronization of laser and ion fields in relatively distant collisions should depend on the ratio of the laser period $T_{\text{field}} = 2\pi/\omega$ and the collision time $T_{\text{coll}} \approx R_0/v_P$ (R_0 characterizes the region in which

transitions are likely). In order to demonstrate that this is indeed the case, the frequency dependence of the results is displayed for a fixed impact velocity in Fig. 2. The field-assisted cross section exhibits a rather broad maximum around $\omega = 0.1$ a.u. for $\delta = 0$, while $\delta = \pi$ suppresses capture strongly; i.e., the (anti-)synchronization is most effective in this region. For $\omega \to 0$, the laser field vanishes in both cases and the field-free cross section is approached. For the phases $\delta = \pi/2, 3\pi/2$, the limit $\omega \to 0$ corresponds to constant electric fields that polarize the electron cloud along and opposite the projectile beam direction such that the total capture cross section is enhanced.

Despite the different behavior of the individual results, the phase-averaged capture is nearly independent of the laser frequency for $\omega \leq 0.2$ a.u. This finding has to be contrasted with some of the earlier molecular calculations that predicted enhanced capture at frequencies close to the relevant quasimolecular energy separations [8]. The insensitivity of our averaged capture cross section with respect to ω indicates that the collision dynamics cannot be predicted from the energy curves of the quasimolecule. From a practical point of view, it permits the use of a variety of different lasers to check the enhancement of the total capture cross section experimentally.

At frequencies around $\omega = 0.3$ a.u., capture is suppressed for all phases δ. A closer analysis of the data shows that a substantial part of the electron density that is captured around the closest approach is removed again from the projectile ion before the propagation of the TDSE is stopped and the analysis of the state vector $|\Psi(t)\rangle$ is performed. It has been checked that pure field ionization of the $He^+(2s)$ and $He^+(2p)$ states, which are dominantly populated in the collision, is rather effective above the two-photon ionization threshold at $\omega = 0.25$ a.u., but not strong enough to explain the observed reduction of the capture cross section. At present, the reason for this reduction is not fully understood.

At higher frequencies which correspond to $T_{\text{field}} \ll T_{\text{coll}}$, the phase dependence of the results becomes negligible. We observe a pronounced peak at $\omega = 0.375$ a.u. and reduced field-assisted capture cross sections above $\omega = 0.5$ a.u., which merge with the field-free result around $\omega = 0.8$ a.u. The peak at $\omega = 0.375$ a.u. originates from a two-step process: First, the $H(2p_0)$ state is excited resonantly by the laser field and exhibits Rabi oscillations with the ground state. In the second step, electrons are transferred very efficiently from the $H(2p_0)$ state to projectile states above the L shell. This process is somewhat distorted by the laser field as one photon is sufficient to ionize the populated states in the outgoing channel.

The same effect is responsible for the reduced field-assisted capture cross section above $\omega = 0.5$ a.u. In this region, electron transfer proceeds similarly to the field-free case around the closest approach, but the dominantly populated $He^+(n = 2)$ states are ionized again before the laser field is turned off and the propagation of the TDSE is stopped. Only at very high frequencies does the field ionization become negligible, since the oscillations are too fast to disturb the electron cloud significantly.

Compared to electron capture, ionization is rather weak at frequencies $\omega < 0.5$ a.u., albeit enhanced with respect to the field-free results. Here, we concentrate on higher ω, for which one-photon ionization is possible and the phase dependence is negligible as in the case of capture. Figure 3 shows the time development of the total electron loss for several frequencies at fixed impact parameter $b = 6$ a.u. The immediate rise of the probabilities at the beginning of the time propagation demonstrates the efficiency of field ionization above the one-photon ionization threshold. However, plateaulike structures are observed before the closest approach except for the highest frequency

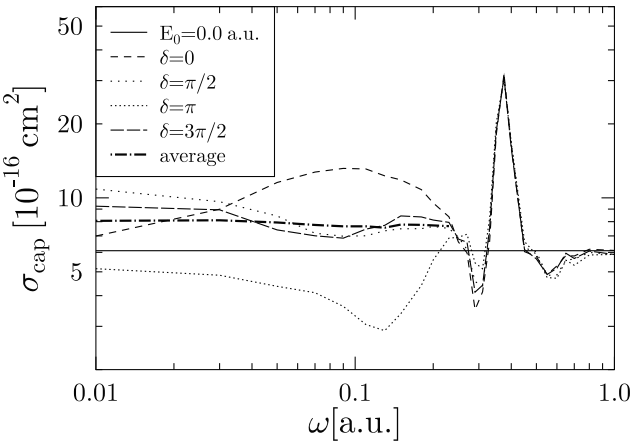

FIG. 2. Total cross sections for electron capture in He^{2+}-H collisions as functions of the laser frequency ω at $E_P = 2$ keV/amu, laser field strength $E_0 = 0.02$ a.u., and longitudinal polarization $\boldsymbol{\epsilon}_{\text{pol}} \| \boldsymbol{v}_P$. Results for different initial phases δ and the phase average are displayed.

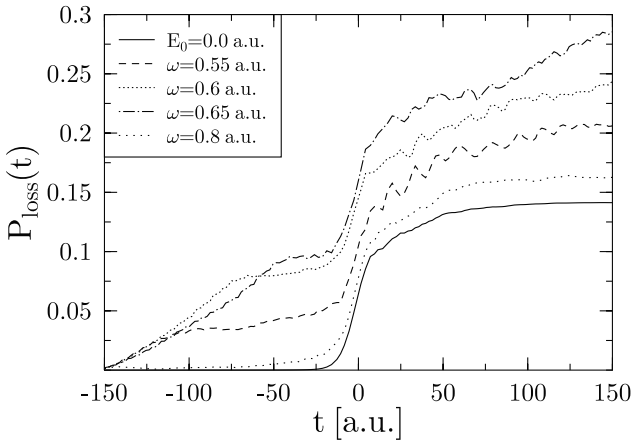

FIG. 3. Time development of total electron loss probabilities for He^{2+}-H collisions at $E_P = 2$ keV/amu, laser field strength $E_0 = 0.02$ a.u., initial phase $\delta = 0$, and longitudinal polarization $\boldsymbol{\epsilon}_{\text{pol}} \| \boldsymbol{v}_P$. Results for different frequencies ω are displayed.

$\omega = 0.8$ a.u. They can be explained by the fact that the binding energy of the initially occupied H($1s$) state $|\varepsilon_{1s}|$ increases as the projectile approaches the target atom [to first order $\varepsilon_{1s}(R) = -0.5 - 2/R$]. When $|\varepsilon_{1s}| > \omega$, one-photon ionization is no longer possible and the electron loss curves flatten. Around the closest approach they rise steeply due to electron capture, and they continue to increase in the outgoing branch as the one-photon ionization channel is opened when the projectile recedes. The ionization process can be viewed as collisionally reduced field ionization. For $\omega = 0.8$ a.u., the reduction is least effective, but as discussed along with Fig. 2 this frequency is too high to produce strong field ionization.

In summary, ionization and capture have been investigated in field-assisted ion-atom collisions. At low frequencies, it was found that capture depends strongly on the initial phase δ, i.e., on the synchronization between laser and projectile interactions. Apparently, one can manipulate the reaction dynamics in ion-atom collisions very efficiently by choosing different phases δ, but this fine-tuning seems unrealizable in an actual experiment. The observable phase-averaged capture cross section is enhanced compared to the field-free result, in particular, at frequencies around the energy separation between the $1s$ and $2p$ target states. In this region, laser-assisted capture proceeds in two steps via the resonant excitation of the H($2p_0$) state. Field ionization is very efficient above the one-photon ionization threshold, but somewhat reduced due to the increased binding of the electron in the presence of the approaching and receding projectile.

These findings can be checked experimentally with sufficiently short laser pulses. The enhanced capture at low frequencies should be observable without such short pulses, since the projectile ions traverse a laser spot focused to a diameter of a few micrometers sufficiently fast to withstand field ionization.

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