Laser-field enhanced electron transfer in *p*-Ne and *p*-Ar collisions

Tom Kirchner*
Institut für Theoretische Physik, TU Clausthal, Leibnizstraße 10, D-38678 Clausthal-Zellerfeld, Germany
(Received 18 October 2006; published 20 February 2007)

Electron transfer in the presence of a laser field is considered for the *p*-Ne and *p*-Ar collision systems. The calculations are based on a time-dependent density functional theory framework and the basis generator method for orbital propagation. Significant enhancements of electron transfer at low collision energy are found and interpreted with the help of molecular potential curves. For the *p*-Ar system a moderately intense field at 800 or 1064 nm wavelength is sufficient to increase the total cross section by more than a factor of 2. Contrary to previously studied cases this scenario can be realized and tested in the laboratory.

DOI: 10.1103/PhysRevA.75.025401 PACS number(s): 34.50.Rk, 32.80.Wr, 34.50.Fa, 34.70.+e

Recent developments in laser technology and multiparticle imaging techniques have opened up new vistas on atomic few-body dynamics [1]. In particular, significant steps have been taken to move from a detailed understanding to steering and, ultimately, active control of such dynamics (see, e.g., Ref. [2] and references therein). Executing control on a femto- or even subfemtosecond time scale is certainly fascinating in itself, but should also be beneficial for a variety of applications.

One scenario of interest is concerned with the simultaneous perturbation of an atomic target by an energetic projectile beam and a strong laser pulse. Experiments are highly challenging and outstanding for the case of ion impact. Nonetheless, there has been considerable theoretical interest in this problem, and a number of recent studies have indeed predicted significant effects—albeit mostly for situations for which experiments are illusory for some time to come. Either the field parameters considered are difficult to achieve, or the collision system involved atomic hydrogen targets, which are delicate to handle in the laboratory and are therefore not favorable for a combined collision and laser-interaction experiment.

Perturbative models were used to study electron transfer [3–5] and ionization [6–9] in fast collisions assisted by relatively weak fields of low frequency, and very recently for ionization in a field with a resonantly tuned higher frequency [10]. A similar tuning at much higher intensities was also studied in terms of a classical simulation [11]. Nonperturbative quantum calculations are computationally costly, and were so far restricted to prototype one-active-electron systems [11–20] and sometimes to reduced-dimensionality models [13,16] or limited basis-set expansions [11] in addition. Particularly clean signatures of the combined action of laser and projectile were found in the electron transfer channel [17,19,20]. For asymmetric collision systems such as He^{2+}-H the field-free cross section is small at low collision velocity, since energy is needed to induce a transition between the relevant quasimolecular states. If, however, the collision is assisted by a field of about $I=1.4\times10^{13}$ W/cm^2 intensity and $\lambda=450$ nm wavelength the cross section is enhanced by more than an order of magnitude below the collision energy $E_P=0.5$ keV/amu [17]. More recent work predicted a less dramatic, but still significant four-to fivefold enhancement at $E_P=0.25$ keV/amu for the more favorable field parameters $I=3.5\times10^{12}$ W/cm^2 and $\lambda=800$ nm, which can be realized by a Ti:sapphire laser [20].

Yet, as mentioned above, these calculations will not be tested experimentally in the near future, since atomic hydrogen is too delicate a target species for a combined collision and laser-interaction experiment. Noble-gas targets are much easier to handle in the laboratory, and are therefore prime candidates for future laser-assisted experiments. These experiments will still be challenging, and attempting them will only be worthwhile if strong effects can be expected. Hence, theoretical predictions for realistic situations are urgently required. To address this need is the purpose of the present communication.

The many-electron nature of the noble-gas atoms complicates the theoretical treatment compared to the (effective) one-electron problems studied previously. Additional approximations, which might influence the accuracy of results are mandatory. Nevertheless, the predictions on significant and observable effects reported below are plausible and are believed to be qualitatively correct for three reasons: (i) our approach is based on time-dependent density functional theory (TDDFT) [21] and proved its reliability in a series of papers for *field-free* collisions (see, e.g., Refs. [22–25]); (ii) the nonperturbative solution of the single-particle equations is accomplished by use of the basis generator method (BGM) [26], whose applicability to laser-assisted collisions has been demonstrated, recently [17]; (iii) the results can be understood by means of qualitative considerations.

The starting point of our calculation is the replacement of the N-electron time-dependent Schrödinger equation that describes the field-assisted collision system in the semiclassical approximation by a set of single-particle equations (in atomic units, i.e., $\hbar=m_e=e=1$)

$$i\partial_t\psi_i(\mathbf{r},t)=\hat{h}(t)\psi_i(\mathbf{r},t),\quad i=1,\ldots,N, \tag{1}$$

with an effective Hamiltonian of the form

$$\hat{h}(t)=-\frac{1}{2}\Delta-\frac{Q_T}{r}-\frac{Q_P}{|\mathbf{r}-\mathbf{R}(t)|}+\mathbf{r}\cdot\mathbf{E}(t)+v_{ee}(\mathbf{r},t). \tag{2}$$

Q_T and Q_P are the charges of the target and the projectile nuclei, respectively, $\mathbf{r}$ is the position vector of an electron

*Electronic address: tom.kirchner@tu-clausthal.de

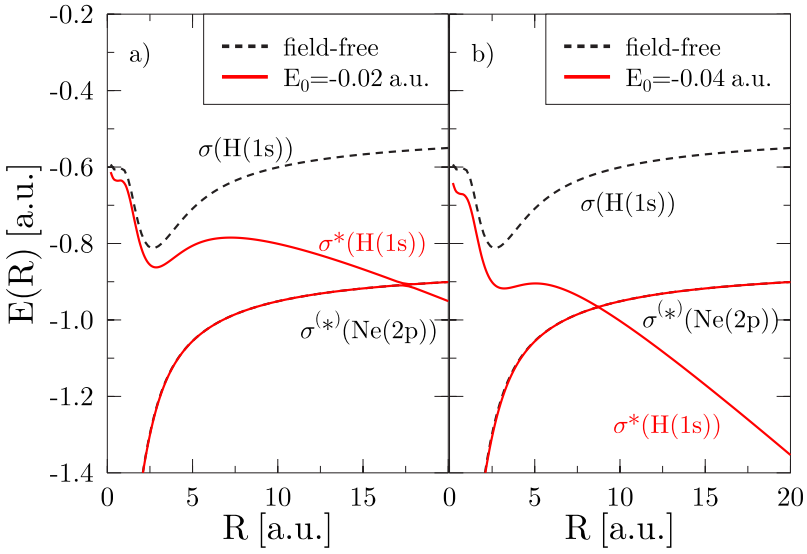

FIG. 1. (Color online) Energy eigenvalues of the $(HNe)^+$ system as functions of the internuclear distance calculated by diagonalizing the single-particle Hamiltonians without field (broken curves) and with static fields (superscript $*$ and full curves) at field strengths E_0= (a) −0.02 and (b) −0.04 a.u. The superscript $(*)$ indicates that the states $\sigma(\mathrm{Ne}(2p))$ and $\sigma^*(\mathrm{Ne}(2p))$ are practically indistinguishable.

with respect to the target center, and $\mathbf{R}(t)=(b,0,vt)$ characterizes the motion of the projectile as a classical straight-line trajectory with impact parameter b and constant velocity v. The laser field in dipole approximation is given by

$$\mathbf{E}(t) = \boldsymbol{\epsilon}_{\mathrm{pol}} E_0 \sin(\omega t + \delta) \tag{3}$$

with constant amplitude E_0, frequency ω, initial phase δ, and a polarization vector $\boldsymbol{\epsilon}_{\mathrm{pol}}$ of unit length, which we assume to be parallel to the ion beam velocity vector (i.e., $\boldsymbol{\epsilon}_{\mathrm{pol}} \| \mathbf{v}$).

The effective potential v_{ee} in Eq. (2) accounts for the electron-electron interaction. According to the basic theorems of TDDFT it is a functional of the one-particle density of the system such that the many-electron problem is mapped exactly to the effective single-particle description. However, its exact form is not known, and appropriate modeling is necessary in practical calculations. Similarly to previous works for field-free p-Ne [22] and p-Ar [24,25] collisions we approximate $v_{ee}(\mathbf{r},t)$ by stationary atomic ground-state potentials $v_{ee}^0(r)$ obtained from the optimized potential method. We comment on the role of response effects, i.e., time-dependent changes in v_{ee} further below.

For both collision systems the single-particle equations (1) have been solved by the nonperturbative BGM, and the same sets of dynamically adapted basis states as in the previous works for field-free collisions have been used. We focus on net electron transfer, which is obtained by summing up the capture contributions (to the *KLM* shells of the hydrogen atom) from all initially populated target orbitals. If one ignores the possible formation of negative hydrogen ions, i.e., multiple capture, net electron transfer can be identified with single-electron transfer [24,25].

In field-free ion-atom collisions at low impact energy electron transfer can be understood by analyzing correlation diagrams of the corresponding quasimolecular systems. The broken curves in Fig. 1 are approximate field-free potential energy curves of the $(HNe)^+$ system obtained from diagonalizing the Hamiltonian $\hat{h}=-\frac{1}{2}\Delta-(Q_T/r)-Q_P/|\mathbf{r}-\mathbf{R}|+v_{ee}^0(r)$ in the BGM basis. From the slopes of the curves one can infer that the coupling between the $\sigma(\mathrm{Ne}(2p))$ and $\sigma(\mathrm{H}(1s))$ states is appreciable at internuclear distances $R \leqslant 5$ a.u. Apparently,

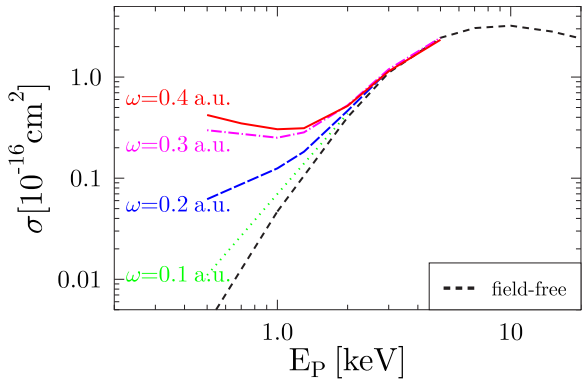

FIG. 2. (Color online) Total electron transfer cross sections as functions of impact energy for p-Ne collisions. BGM results for field-free and phase-averaged laser-assisted collisions at field strength E_0=0.02 a.u., different frequencies ω, and longitudinal polarization $\boldsymbol{\epsilon}_{\mathrm{pol}} \| \mathbf{v}$.

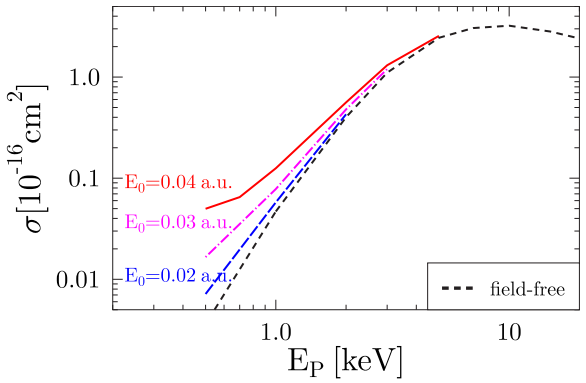

FIG. 3. (Color online) Total electron transfer cross sections as functions of impact energy for p-Ne collisions. BGM results for field-free and phase-averaged laser-assisted collisions at field frequency ω=0.057 a.u., different field amplitudes E_0, and longitudinal polarization $\boldsymbol{\epsilon}_{\mathrm{pol}} \| \mathbf{v}$.

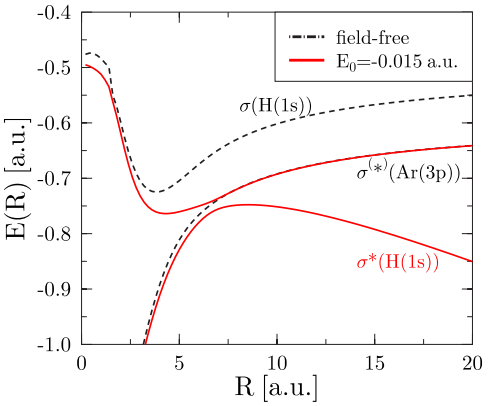

FIG. 4. (Color online) Energy eigenvalues of the $(HAr)^+$ system as functions of the internuclear distance calculated by diagonalizing the single-particle Hamiltonians without field (broken curves) and with a static field (superscript $*$ and full curves) at field strength $E_0=-0.015$ a.u. The superscript $(*)$ indicates that $\sigma(\mathrm{Ar}(3p))$ and $\sigma^*(\mathrm{Ar}(3p))$ are practically indistinguishable at large internuclear distance.

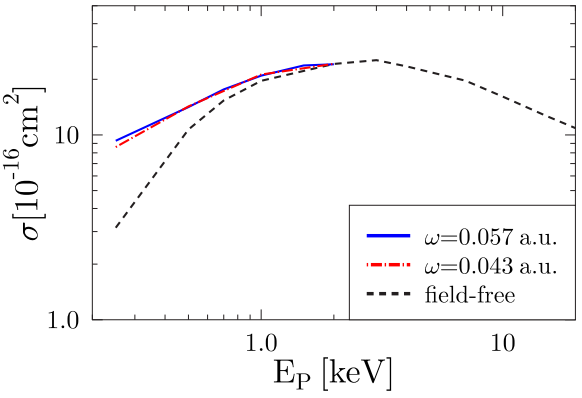

FIG. 5. (Color online) Total electron transfer cross sections as functions of impact energy for p-Ar collisions. BGM results for field-free and phase-averaged laser-assisted collisions at field strength $E_0=0.015$ a.u., different frequencies ω, and longitudinal polarization $\boldsymbol{\epsilon}_{\mathrm{pol}} \| \mathbf{v}$.

about 0.3 a.u. energy is needed for the corresponding transition. In very slow collisions it is unlikely that the energy is transferred from the heavy-particle motion, which is why the field-free transfer cross section is small in this region (Fig. 2).

It was argued in earlier works for one-electron systems that field-assisted transfer can nevertheless be significant if the laser frequency is tuned such that it matches the energy difference between the quasimolecular states in the coupling region [17,27]. In order to test this idea we have performed transfer calculations for the p-Ne system for frequencies from $\omega=0.1$ to 0.4 a.u. at the same field strength as in our previous work for He^{2+}-H collisions, $E_0=0.02$ a.u. ($I=1.4\times10^{13}$ W/cm^2). Results of these calculations are shown in Fig. 2. Indeed, the total electron transfer cross section is enhanced dramatically at low impact energy for $\omega=0.3$ and 0.4 a.u., while lower frequencies are less efficient.

The frequency $\omega=0.3$ a.u. corresponds to a wavelength of about 150 nm. While it is not impossible to generate and use such a field, it would be much more convenient if a laser-assisted electron transfer experiment could be done with the fundamentals of standard Ti:sapphire or neodymium-doped yttrium aluminum garnet (Nd:YAG) lasers at $\lambda=800$ nm ($\omega=0.057$ a.u.) or $\lambda=1064$ nm ($\omega=0.043$ a.u.), respectively. In these cases the field oscillations are relatively slow compared to the collision time, which suggests that an appropriate physical picture emerges from studying the potential curves of the quasimolecule dressed by a static field [20]. The field bends the potential curves, and if crossings occur in regions in which states are coupled, significant transitions can be expected. Accordingly, we have also diagonalized the single-particle Hamiltonian including the term that describes the interaction with the field. Results for $E_0=-0.02$ and -0.04 a.u. are shown as full curves in Fig. 1. The sign is chosen such that the electric field points from the proton to the Ne center. Indeed, crossings of the states in question do occur. However, only for the higher field strength is this crossing located at an internuclear distance small enough to enhance electron transfer significantly.

This is confirmed by Fig. 3, which displays total cross sections for electron transfer for the wavelength $\lambda=800$ nm. While the field strength $E_0=0.02$ a.u. is rather inefficient, $E_0=0.03$ a.u., and, in particular, $E_0=0.04$ a.u. lead to strong enhancements of electron transfer below the collision energy $E_P=1$ keV. Even stronger fields would move the crossing of the potential curves below $R=5$ a.u., i.e., into the region, in which the collisional coupling maximizes. However, $E_0=0.04$ a.u. corresponds already to a rather high intensity ($I=5.6\times10^{13}$ W/cm^2), such that field ionization of the target or the projectile after capture becomes an issue. On the time scale of the collision the total ionization probabilities are still well below 1% according to the present calculations, but in an experiment it will be difficult to extract a clean signal for laser-assisted electron transfer if field ionization can occur before and after the collision.

These considerations imply that the p-Ne system is probably not the ideal first candidate for such an experiment. Instead, a collision system with a smaller energy splitting of the field-free potential curves would be favorable, since a field-induced crossing at not too large internuclear distance would then occur at lower field strength. Figure 4 shows that the p-Ar system is such a candidate. In this case the moderate field strength $E_0=0.015$ a.u. ($I\approx8\times10^{12}$ W/cm^2) induces an avoided crossing at $R\approx6$ a.u. between the two σ states whose coupling gives rise to electron transfer.

Again, we have performed calculations for electron transfer in this system and present total cross sections for both the Ti:sapphire and the Nd:YAG wavelengths in Fig. 5. They are very similar and significantly enhanced compared to the field-free result at low impact energy.[1] We have checked that also on the level of impact-parameter-dependent transition

[1]Even at $E_P=0.25$ keV the straight-line approximation is expected to be reliable: the main contributions to the total cross section come from impact parameters $1<b<6$ a.u. Assuming $N-1$ passive electrons the repulsion of projectile and target can be estimated by classical Rutherford scattering with the charge product $Q_PQ_T=1$. The distance of closest approach then amounts to $R_{\mathrm{min}}\approx b+0.055$ a.u. for $b\geqslant1$ a.u..

probabilities do both wavelengths give almost identical results. This adds to the reliability of the static field interpretation, as it shows that the time dependence of the fields has no major influence.

As mentioned earlier the present calculations do not account for response effects in the effective potential v_{ee} that appears in the Hamiltonian (2). We have estimated these effects based on the approximate response model introduced in Ref. [23]. While they turned out to be insignificant for the p-Ne system, they reduce the p-Ar cross sections by 20–30 %. However, the enhancement of the field-assisted compared to the field-free electron transfer cross section is not significantly affected, but only slightly reduced from a factor of 2.7 to a factor of 2.5 at the lowest impact energy considered, i.e., it should still be observable in an experiment.

Summarizing, we have presented quantum-mechanical calculations for laser-assisted electron transfer in p-Ne and p-Ar collisions. In both cases, significant enhancements of the total cross sections at low impact energy have been observed and interpreted by means of potential curves of the quasimolecular systems. At relatively high field frequency and low intensity an energy matching argument is able to explain the enhancement of electron transfer in p-Ne collisions. For fields conveniently generated by standard Ti:sapphire or Nd:YAG lasers curve crossings of molecular states dressed by static fields mirror possible enhancements. While rather strong fields are needed in the p-Ne case, the moderate intensity $I \approx 8 \times 10^{12}$ W/cm^2 is sufficient to increase the p-Ar cross section by more than a factor of 2 at low impact energy. In contrast to previous predictions this result can be tested experimentally with present-day technology: the field and collision parameters are realizable, and argon atoms are easy to handle in the laboratory.

Finally, a few comments are in order with regard to the experimental situation for field-free electron transfer. To the author's knowledge only a few rather old experimental data sets are available for the situations of interest. They have not been included in the figures so as not to obscure the comparison with field-assisted transfer. In the p-Ne case the experimental total cross sections reported by Allison [28] are significantly larger than our calculations at $E_P \leqslant 2$ keV. However, very recent measurements are in much better agreement with the latter [29]. For field-free p-Ar collisions we have discussed the comparison with experimental data down to $E_P = 1$ keV in Ref. [25]. In this case also an experimental reexamination would be of interest to assess the situation on a more quantitative level. Because of the arguments given above it is believed that the reported enhancement of electron transfer in the presence of a laser field is not significantly affected by these uncertainties. Ultimately, this can only be decided by experimental investigations.

I thank T. Spranger and L. F. Menchero for assistance, and H. Tawara for the communication of yet unpublished data on field-free p-Ne collisions. This work has been supported by the Deutsche Forschungsgemeinschaft.

[1] *Many-Particle Quantum Dynamics in Atomic and Molecular Fragmentation*, edited by J. Ullrich and V. P. Shevelko (Springer, Heidelberg, 2003).
[2] M. Y. Ivanov, R. Kienberger, A. Scrinzi, and D. M. Villeneuve, J. Phys. B **39**, R1 (2006).
[3] S.-M. Li, Y.-G. Miao, Z.-F. Zhou, J. Chen, and Y.-Y. Liu, Z. Phys. D: At., Mol. Clusters **39**, 29 (1997).
[4] S.-M. Li, X.-L. Xu, Z.-F. Zhou, J. Chen, and Y.-Y. Liu, Eur. Phys. J. D **2**, 237 (1998).
[5] S.-M. Li, Y.-G. Miao, Z.-F. Zhou, J. Chen, and Y.-Y. Liu, Phys. Rev. A **57**, 3705 (1998).
[6] A. B. Voitkiv and J. Ullrich, J. Phys. B **34**, 1673 (2001).
[7] A. B. Voitkiv and J. Ullrich, J. Phys. B **34**, 4383 (2001).
[8] S.-M. Li, J. Chen, and Z.-F. Zhou, J. Phys. B **35**, 557 (2002).
[9] M. F. Ciappina and L. B. Madsen, J. Phys. B **39**, 5037 (2006).
[10] A. B. Voitkiv, N. Toshima, and J. Ullrich, J. Phys. B **39**, 3791 (2006).
[11] L. B. Madsen, J. P. Hansen, and L. Kocbach, Phys. Rev. Lett. **89**, 093202 (2002).
[12] T. Kirchner, Phys. Rev. Lett. **89**, 093203 (2002).
[13] M. Lein and J. M. Rost, Phys. Rev. Lett. **91**, 243901 (2003).
[14] J. P. Hansen, T. Sørevik, and L. B. Madsen, Phys. Rev. A **68**, 031401(R) (2003).
[15] M. S. Pindzola, T. Minami, and D. R. Schultz, Phys. Rev. A **68**, 013404 (2003).
[16] T. Niederhausen, B. Feuerstein, and U. Thumm, Phys. Rev. A **70**, 023408 (2004).
[17] T. Kirchner, Phys. Rev. A **69**, 063412 (2004).
[18] T. Kirchner, Nucl. Instrum. Methods Phys. Res. B **233**, 151 (2005).
[19] T. Niederhausen and U. Thumm, Phys. Rev. A **73**, 041404(R) (2006).
[20] F. Anis, V. Roudnev, R. Cabrera-Trujillo, and B. D. Esry, Phys. Rev. A **73**, 043414 (2006).
[21] *Time-Dependent Density Functional Theory*, edited by M. A. L. Marques, C. A. Ullrich, F. Nogueira, A. Rubio, K. Burke, and E. K. U. Gross, Lecture Notes in Physics Vol. 706 (Springer, Heidelberg, 2006).
[22] T. Kirchner, H. J. Lüdde, and R. M. Dreizler, Phys. Rev. A **61**, 012705 (2000).
[23] T. Kirchner, M. Horbatsch, H. J. Lüdde, and R. M. Dreizler, Phys. Rev. A **62**, 042704 (2000).
[24] T. Kirchner, M. Horbatsch, and H. J. Lüdde, Phys. Rev. A **66**, 052719 (2002).
[25] T. Kirchner, M. Horbatsch, M. Keim, and H. J. Lüdde, Phys. Rev. A **69**, 012708 (2004).
[26] O. J. Kroneisen, H. J. Lüdde, T. Kirchner, and R. M. Dreizler, J. Phys. A **32**, 2141 (1999).
[27] L. F. Errea, L. Méndez, and A. Riera, J. Chem. Phys. **79**, 4221 (1983).
[28] S. K. Allison, Rev. Mod. Phys. **30**, 1137 (1958).
[29] T. Kusakabe, H. Tawara, M. Kimura, J. P. Gu, and R. J. Buenker (unpublished).