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Electron emission during combined attosecond pulses

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When a strong attosecond laser pulse acts simultaneously with a fast ionic projectile, the electronic response of an atom changes and leads to a drastic change in the final momenta, which may be observed in recoil-ion momentum experiments. The nonlinear response to the combined fields leads in certain cases to ionization probabilities more than three times larger than the probabilities by either of the two fields individually. The results are based on an algorithm for accurate solution of the time-dependent Schrödinger equation in three dimensions, which has the property that it requires CPU time comparable with that of two-dimensional methods.

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As attosecond laser pulses with full phase control are becoming reality [1], a number of electronic processes are likely to be studied in the time domain. For example, processes such as Auger decay [2] and collision-induced charge transfer [3] can in principle be monitored with assisting attosecond ''cameras'' [4] to gain new insight into the detailed electron dynamics. Along these lines it is also interesting to bear in mind that exposing any electronic system to several perturbations may lead to interference effects, testing the processes at the level of phases, and resulting in strongly altered dynamics.

In a recent work [5] it was suggested that the dynamics of a collision-induced continuum electron would be drastically changed by the presence of an attosecond laser pulse. The final electron momenta would carry a signature of precisely how the laser pulse and the projectile Coulomb interaction worked with respect to each other, and it should be possible to observe the effects by recoil-ion momentum measurements [6]. The conclusion in Ref. [5] was, however, based on classical Monte Carlo calculations. To include possible coherent dynamics and interference the need for *ab initio* quantum-mechanical calculations is evident. Such calculations are complex since the lack of symmetry requires a full three-dimensional solution of the time-dependent Schrödinger equation (TDSE). Standard numerical methods for laser-atom interactions rely on the conservation of, e.g., the magnetic quantum number m in linearly polarized fields [7,8], and such schemes then become inapplicable. Also recent advanced three-dimensional methods are either specialized towards atomic collision processes [9–11] or laser-atom interactions [12], only. This motivated us to develop a flexible method applicable for a general time-dependent field. Hence, the purpose of the present work is twofold. First, a powerful and very general numerical method will be presented. Second, fully quantum-mechanical calculations, only practical to carry out with the method, address the question of collisions in combined fields of attosecond duration and it is investigated to which extent the electron dynamics driven by laser pulses *and* fast ions is altered compared to ''laser only'' or to ''collision only'' processes.

The method we propose solves the TDSE on a grid consisting of a finite set of points representing three-dimensional space. On the grid, the dynamical equations are propagated by a split operator [13]. In Cartesian coordinates the spectral split-operator method is readily implemented and many applications have been found, but these coordinates are often inconvenient since they do not allow the introduction of any underlying symmetry pertaining to the problem at hand. In contrast, the core of our method is to expand the wave function in orthogonal polynomials which diagonalize part of the Hamiltonian effectively. For the processes we are interested in here, we take advantage of the spherical symmetry of the atomic potential $V_0(r)$ and express the TDSE in spherical coordinates for the scaled wave function $\Phi = r\Psi$ [atomic units (a.u.) are used throughout]:

$$\left(-\frac{1}{2}\frac{\partial^2}{\partial r^2} + \frac{\boldsymbol{L}^2}{2r^2} + V_0(r) + W(\boldsymbol{r},t) - i\frac{\partial}{\partial t} \right) \Phi = 0, \quad (1)$$

where $W(\boldsymbol{r},t)$ is a general perturbation depending on time and space. In the present application we consider the hydrogen atom, $V_0(r) = -1/r$, but the method will work for any potential favoring a representation in spherical coordinates. Introducing $A = -\frac{1}{2}\partial^2/\partial r^2$ and $B = \boldsymbol{L}^2/2r^2 + V_0(r)$, the wave function at time $t + \Delta t$, with Δt small, can be evaluated in the split step form [13,14]

$$\Phi(\boldsymbol{r},t+\Delta t) = e^{-i\Delta t A/2} e^{-i\Delta t B/2} e^{-i\Delta t W(\boldsymbol{r},t)} e^{-i\Delta t B/2} e^{-i\Delta t A/2} \Phi \times (\boldsymbol{r},t) + \mathcal{O}(\Delta t^3). \quad (2)$$

The error term represents the splitting error and it disappears when the operators commute. The B operator combines more commuting operators, thus evaluation of B can be carried out in individual steps without further splitting errors.

An efficient and accurate numerical scheme is obtained by expanding $\Phi(\boldsymbol{r},t)$ in spherical harmonics $Y_{lm}(\Omega_{jk})$ defined on a finite number of points $\Omega_{jk} = (\theta_j, \phi_k)$,

$$\Phi(r_i, \Omega_{jk}, t) = \sum_{l,m}^{l_{\max}} f_l^m(r_i,t) Y_{lm}(\Omega_{jk}), \quad (3)$$

where r_i defines the radial grid points. For fixed m (fixed ϕ_k), the discrete versions of the spherical harmonics maintain their unitarity property if the θ_j's in $\Omega_{jk}=(\theta_j,\phi_k)$ are chosen as the Gauss-Jacobi quadrature points [14]. However, when the azimuthal symmetry is broken m is not conserved, and no known two-dimensional analog exists and in fact the Gauss-Jacobi points and weights depend on m, which again makes the method unattractive. The way out of this dilemma from related mathematical physics of, e.g., methods of geophysical research [15] has been to reexpand the spherical harmonics in a basis of associated Legendre polynomials in θ and a Fourier basis in ϕ on a regular grid. A consequence of this is a clustering of evaluation points near the poles, which implies several undesirable features. These can be eliminated by adding clever ''filtering'' technique, with the unavoidable consequence of a rather complicated code.

Alternatively one may take any interpolatory quadrature rule over the sphere which integrates exactly all polynomials of degree $\leqslant 2l_{\max}$. Recently abscissas and weights for such rules were computed [16,17] and made available over the Internet [18]. The points are distributed almost equidistant, thus no ''pole problem'' exists and for a specific $l_{\max}$, the number of angular points is $(2l_{\max}+1)^2$. In the calculations we have used $l_{\max}=7$ and on the radial grid with radius $R_0=100$ we have used 2048 points, and in the propagation we have used $\Delta t=0.01$. We have integrated until $vt=60$, with $v=2$ the velocity of the proton projectile. Tests for convergence were performed with 4096 radial points and $l_{\max}=15$.

Taking Ω_{jk},w_{jk} as the set of abscissas and weights ensures that the standard orthogonality property is fulfilled,

$$\delta_{l,l'}\delta_{m,m'}=\sum_{jk} w_{jk}Y^*_{l'm'}(\Omega_{jk})Y_{lm}(\Omega_{jk}). \tag{4}$$

The radial basis functions can at any time be constructed by

$$f^m_l(r_i,t)=\sum_{jk} w_{jk}Y^*_{l'm'}(\Omega_{jk})\Phi(r_i,\Omega_{jk},t), \tag{5}$$

which is a discrete version of textbook projection theory of quantum mechanics. The application of the time propagator (2) involves first that each radial function $f^m_l(r_i,t)$ is represented in momentum space,

$$f^m_l(r_i,t)=\sum_k g^m_{k,l}e^{i(\pi/R_0)kr_i}. \tag{6}$$

The kinetic-energy operator results in a multiplicative factor onto each Fourier coefficient, i.e., $g^m_{k,l}\rightarrow e^{i(\pi/R_0)^2k^2/2}g^m_{k,l}$. The inverse Fourier transform is applied, and the effect of the angular-momentum operator is a multiplicative factor, $e^{-i\Delta tB}f^m_l(r_i,t)\rightarrow e^{-i\Delta t[l(l+1)/2r_i^2+V_0(r_i)]}f^m_l(r_i,t)$. Finally, the total wave function is reconstructed as in Eq. (3) and the effect of the external potential is evaluated by $\Phi(r_i,\Omega_{jk},t+\Delta t)=e^{-i\Delta tW(\mathbf{r},t)}\Phi(r_i,\Omega_{jk},t)$. These steps are repeated in appropriate order to propagate the wave function. The initial state may be found by propagation in imaginary time.

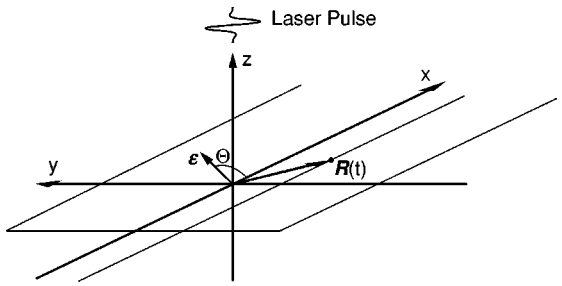

FIG. 1. Collision geometry. The collision plane (xy) is defined by the projectile trajectory $\boldsymbol{R}(t)$. The laser is propagating perpendicular to the scattering plane with an in-plane linear polarization vector $\boldsymbol{\epsilon}=(\cos\Theta,\sin\Theta)$. In the calculations we consider p-H$(1s)$ collisions with impact parameter $\boldsymbol{b}=3\hat{\boldsymbol{e}}_y$ and velocity $\boldsymbol{v}=2\hat{\boldsymbol{e}}_x$. The laser is modeled by a sinusoidal pulse with a $\sin^2$ envelope of duration $\tau=16$. See text for details.

With the central role of spherical harmonics in many areas of quantum dynamics, the practical implementation of Eqs. (2)–(6) in computer codes cannot be overestimated. The advantage of the present method is foremost that relatively few points Ω_{jk} make the method quasi-two-dimensional when high values of $l_{\max}$ are not generated by the external potential $W(\boldsymbol{r},t)$. Keeping the total number of numerical mesh points constant, the present method thus allows for a much larger radial mesh compared to, e.g., Cartesian grid points. A larger spatial volume may thus be discretized at the same computational cost. The Fourier transform as well as the propagating potential can be performed in parallel [19] with a minimum of communication time.

The method is therefore ideal for a range of time-dependent phenomena such as vortex dynamics in Bose-Einstein condensates [20], atoms and artificial atoms [21] in time-dependent electromagnetic fields, as well as ion-atom collisions. For electron emission in particular, the outgoing Coulomb wave is obtained by a direct radial one-dimensional integration and the differential cross section in a certain direction is given by

$$\frac{dP}{d\boldsymbol{k}}\propto\left|\sum_{l,m}(-i)^l e^{i\delta_l}Y_{lm}(\hat{\boldsymbol{k}})\int_0^{R_0}drrR_{k,l}(r)f^m_l(r)\right|^2, \tag{7}$$

where δ_l is the phase shift of the lth partial wave and $R_{k,l}(r)$ is a radial continuum function. The exact total ionization probability can be evaluated by performing the radial projection onto all positive k states which vanish at R_0 or, when capture can be neglected, by subtracting the sum of all populated excited states, $1-\Sigma_{n,l}\int_0^{R_0}drrR_{nl}(r)f^m_l(r)$. The continuum-electron analysis discussed here is more complicated on a Cartesian grid where the angular behavior cannot be factored out.

Consider now the physical situation shown in Fig. 1. A collision plane is identified by the projectile trajectory $\boldsymbol{R}(t)=\boldsymbol{v}t+\boldsymbol{b}$ with $\boldsymbol{v}$ the velocity and $\boldsymbol{b}$ the impact parameter, which can be detected by coincidence measurements [22]. The laser propagates perpendicular to the collision plane and has an in-plane linear polarization vector $\boldsymbol{\epsilon}=(\cos\Theta,\sin\Theta)$ where Θ is the angle with respect to the x axis. The corre-

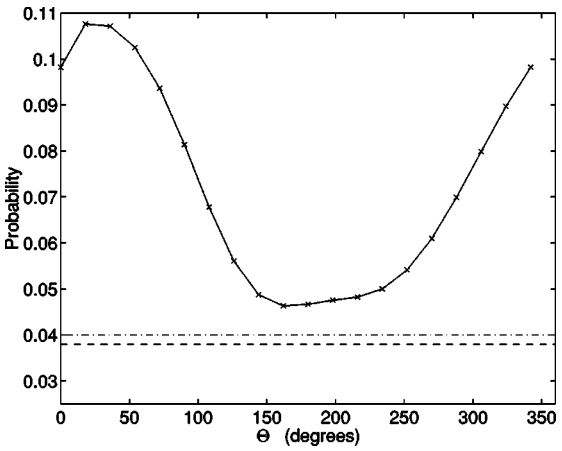

FIG. 2. Ionization probability in p-H($1s$) collisions as a function of the direction of the in-plane laser polarization vector at $\boldsymbol{b}=3\hat{\boldsymbol{e}}_y$ and $\boldsymbol{v}=2\hat{\boldsymbol{e}}_x$. The collision only probability (dashed-dotted line) and the laser only ionization probability (dashed line) are marked as straight lines. See text for the parameters of the laser pulse.

sponding interaction part of the Hamiltonian reads $W(\boldsymbol{r},t)=-Z_p/|\boldsymbol{R}(t)-\boldsymbol{r}|-E(t)[x\cos\Theta+y\sin\Theta]$, with $E(t)=f_L(t)E_0\cos(\omega t-\delta)$ the laser field, E_0 the field strength, $f_L(t)$ the pulse shape, ω the laser frequency, and δ the phase of the field, adjusted to secure a zero dc field component after the end of the pulse [23]. In the calculations, we use a finite $\sin^2$ pulse, $f_L(t)=\sin^2\pi(t/\tau-1/2)$ for $-\tau/2\leqslant t\leqslant\tau/2$, $\tau=16$ and $f_L(t)=0$ otherwise, $E_0=0.1$ and $\omega=0.375$ corresponding to two-photon ionization. The Keldysh parameter of $\gamma=3.75$ places the study in the multiphoton ionization regime (tunneling ionization corresponds to $\gamma\ll1$) [24].

We focus at an impact velocity of the proton which is sufficiently high to minimize capture, $\boldsymbol{v}=2\hat{\boldsymbol{e}}_x$. The impact parameter $\boldsymbol{b}=3\hat{\boldsymbol{e}}_y$ was chosen because our previous study [5] indicated pronounced interference effects between excitation induced by the projectile and excitation induced by the laser for impact parameters between 1 and 7 a.u.

Figure 2 displays the ionization probability as a function of the angle Θ. A strong dependence on the polarization direction is observed. When the laser polarization is parallel with the projectile velocity ($\Theta=0°$), the probability for electron emission in a combined process is seen to be close to a factor of 3 larger than if any of the ionization processes acts isolated. Furthermore, there is a backward-forward asymmetry of the probability which is intimately linked to the phase of the laser pulse. In the backward direction the effect of the two fields adds up to an emission probability only slightly larger than the single-field probability. In both directions, only a strong interference effect between the two perturbations can cause this result.

We now address the suggestion that the present attosecond interference phenomena might be measured in recoil measurements [5]. The impact parameter is assumed fixed and detected by the ''ion'' detector. The question is then whether the electron signal (momenta) carries information on which type of process took place, i.e., laser only, collision

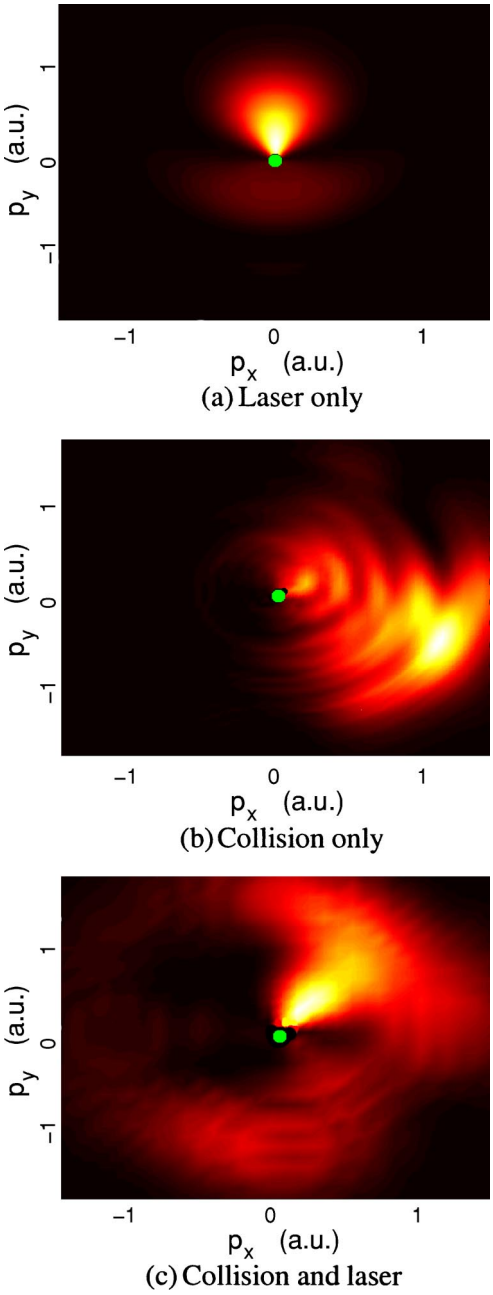

FIG. 3. Differential cross section of the ejected electron momenta in the collision plane for ionization in p-H($1s$) for $\boldsymbol{b}=3\hat{\boldsymbol{e}}_y$ and $\boldsymbol{v}=2\hat{\boldsymbol{e}}_x$ and polarization vector with $\Theta=90°$. See text for the parameters of the laser pulse. The origin is highlighted by a bullet.

only, or a mixed type. In Fig. 3 the final electron momenta in the collision plane are plotted in the target frame. It is seen that the laser only and collision only processes remain separated: the laser pulse (at $\Theta=90°$, corresponding to a polarization vector perpendicular to the impact parameter) ionizes predominantly along the positive y axis. Asymmetric photoelectron emission is well known in few-cycle fields [25,26] and was recently measured and related to the carrier-envelope phase difference [27,28].

The collision only process drags the electron towards the

projectile, and the electrons tend to have a large positive x component peaked at longitudinal momenta (p_x) of half the projectile momentum, and with a peak at negative transverse momenta. This confirms previous experimental [29–31] and theoretical [9,31] studies. What is important in Fig. 3(b) is the detailed diffractionlike pattern at the backside of the scatterer.

The fact that the two processes in the emission spectrum are rather isolated opens the possibility for electron emission in combined fields to take place with its own characteristic final momenta. In Fig. 3(c) this is precisely what happens: the strong y component of the momentum of the laser emission is combined with a strong positive x component from the collision resulting in electrons arriving with both positive x and y components. Thus under certain ideal experimental conditions the electron momenta in combination with controlled impact parameters allow for a trace back and measurement of sophisticated interference effects on the attosecond time scale.

In summary, we have shown the results of a spectral algorithm which allow for full advantage of parallel computers, to the solution of the TDSE in three dimensions. The general idea of the method is to use coordinates accounting for part of the symmetry in the problem in combination with advanced quadrature rules. Based on its generality we foresee implementations in many branches of physics, and here, as an example of current interest, we have used the computational algorithm to show that electron emission momenta in strong combined attosecond fields carry information on which type of attosecond process was working.

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[1] A. Baltuška, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V.S. Yakovlev, T.W.H.A. Scrinzi, and F. Krausz, Nature (London) **421**, 611 (2003).
[2] M. Drescher, M. Hentschal, R. Kienberger, M. Uiberacker, V. Yakovlev, A. Scrinzi, T. Westerwalbasloh, U. Kleineberg, U. Heinzmann, and F. Krausz, Nature (London) **419**, 803 (2001).
[3] T. Kirchner, Phys. Rev. Lett. **89**, 093203 (2002).
[4] M. Hentschel, R. Kienberger, C. Spielmann, G.A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature (London) **414**, 509 (2001).
[5] L.B. Madsen, J.P. Hansen, and L. Kocbach, Phys. Rev. Lett. **89**, 093202 (2002).
[6] R. Dörner, V. Mergel, O. Jagutzki, L. Spielberger, J. Ullrich, R. Moshammer, and H. Schmidt-Böcking, Phys. Rep. **330**, 95 (2000).
[7] P. Lambropoulos, P. Maragakis, and J. Zhang, Phys. Rep. **305**, 203 (1998).
[8] D. Dundas, Phys. Rev. A **65**, 023408 (2002).
[9] E.Y. Sidky, C. Illescas, and C.D. Lin, Phys. Rev. Lett. **85**, 1634 (2000).
[10] J.C. Wells, D.R. Schultz, P. Gavras, and M.S. Pindzola, Phys. Rev. A **54**, 593 (1996).
[11] M.E. Riley and B. Ritchie, Phys. Rev. A **59**, 3544 (1999).
[12] D.-I. Choi and W. Chism, Phys. Rev. A **66**, 025401 (2002).
[13] M.D. Feit, J.A. Fleck, and A. Steiger, J. Comput. Phys. **47**, 412 (1982).
[14] M.R. Hermann and J.A. Fleck, Phys. Rev. A **38**, 6000 (1988).
[15] P. Swarztrauber, SIAM (Soc. Ind. Appl. Math.) J. Numer. Anal. **16**, 934 (1979).
[16] R.S. Womersley and I.H. Sloan, Adv. Comput. Math. **14**, 195 (2001).
[17] I.H. Sloan and R.S. Womersley, Adv. Comput. Math. (to be published).
[18] I.H. Sloan and R.S. Womersley, http://www.maths.unsw.edu.au/rsw/Sphere/.
[19] J.P. Hansen, T. Matthey, and T. Sørevik, in *Recent Advances in Parallel Virual Machine and Message Passing Interface. 10th European PVM/MPI Users' Group Meeting*, Lecture Notes in Computer Science (Springer-Verlag, Heidelberg, in press).
[20] D. McPeake, H.M. Nilsen, and J.F. McCann, Phys. Rev. A **65**, 063601 (2002).
[21] G.Y. Slepyan, S.A. Maksimenko, A. Hoffmann, and D. Bimberg, Phys. Rev. A **66**, 063804 (2002).
[22] M. van der Poel, C.V. Nielsen, M.-A. Gearba, and N. Andersen, Phys. Rev. Lett. **87**, 123201 (2001).
[23] L.B. Madsen, Phys. Rev. A **65**, 053417 (2002).
[24] L.V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].
[25] E. Cormier and P. Lambropoulos, Eur. Phys. J. D **2**, 15 (1998).
[26] J.P. Hansen, J. Lu, L.B. Madsen, and H.M. Nilsen, Phys. Rev. A **64**, 033418 (2001).
[27] G.G. Paulus, F. Grasbon, H. Walther, P. Villoresi, M. Nisoli, S. Stagira, E. Priori, and S.D. Silvestri, Nature (London) **414**, 182 (2001).
[28] D.B. Milošević, G.G. Paulus, and W. Becker, Phys. Rev. Lett. **89**, 153001 (2002).
[29] R. Dörner, H. Khemliche, M.H. Prior, C.L. Cocke, J.A. Gary, R.E. Olson, V. Mergel, J. Ullrich, and H. Schmidt-Böcking, Phys. Rev. Lett. **77**, 4520 (1996).
[30] Th. Weber, Kh. Khayyat, R. Dörner, V.D. Rodriguez, V. Mergel, O. Jagutzki, L. Schmidt, K.A. Müller, F. Afaneh, A. Gonzalez, and H. Schmidt-Böcking, Phys. Rev. Lett. **86**, 224 (2001).
[31] E. Edgü-Fry, C.L. Cocke, E. Sidky, C.D. Lin, and M. Abdallah, J. Phys. B **35**, 2603 (2002).