

CHCRUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Exploration of the subcycle multiphoton ionization dynamics and transient electron density structures with Bohmian trajectories

Hossein Z. Jooya, Dmitry A. Telnov, Peng-Cheng Li, and Shih-I Chu Phys. Rev. A **91**, 063412 — Published 18 June 2015 DOI: 10.1103/PhysRevA.91.063412

Exploration of the sub-cycle multiphoton ionization dynamics and transient electron density 1 2 structures with Bohmian trajectories 3 Hossein Z. Joova^{1,*}, Dmitry A. Telnov², Peng-Cheng Li^{3,4}, and Shih-I Chu^{1,3,*} 4 5 ¹Department of Chemistry, University of Kansas, Lawrence, Kansas 66045, USA 6 ²Department of Physics, St. Petersburg State University, St.Petersburg 198504, Russia 7 ³Center for Quantum Science and Engineering, Department of Physics, National Taiwan 8 9 University, Taipei 10617, Taiwan ⁴College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou, Gansu 10 730070, China 11 12 * jooya@ku.edu , sichu@ku.edu 13 14 PACS numbers: 32.80.Rm,42.50.Hz 15 16 17 18 Abstract 19 An accurate 3D numerical scheme for the De Broglie-Bohm's framework of Bohmian mechanics is presented. This method is utilized to explore the sub-cycle multiphoton ionization 20 dynamics of the hydrogen atom subject to intense near infrared (NIR) laser fields on the sub-21 femtosecond time scale. The analysis of the time-dependent electron density reveals that several 22 distinct density portions can be shaped and detached from the core within a half cycle of the laser 23 field. As a complementary perspective, we identify several distinct groups of the Bohmian 24 trajectories which represent the multiple detachments of the electron density at different times. 25 The method presented provides very accurate electron densities and Bohmian trajectories that 26 27 allow to uncover the origin of the formation of the transient and distinct electron structures seen in the MPI processes. 28 29 30 31 The recent development of attosecond metrology has enabled the real-time experimental 32

observation of ultrafast electron dynamics in atomic and molecular systems [1,2]. Considerable interest has been recently paid also to the study of transient absorption spectroscopy in ultrafast 33 time domain [3-5]. For example, the observation of the transient changes in the absorption of an 34 isolated attosecond XUV pulse by helium atoms in the presence of a delayed few-cycle NIR 35 36 laser pulse has been recently reported and uncovered novel absorption structures corresponding to laser-induced "virtual" intermediate states in the two-color two-photon (XUV + NIR) and 37 38 three photon (XUV+ NIR + NIR) absorption processes [5]. These previously unobserved absorption structures are modulated on half-cycle (~ 1.3 fs) and quarter-cycle (~ 0.6 fs) time 39 40 scales, resulting from quantum optical interference in the laser-driven atom. More recently, there is also new interest in the study of sub-cycle transient high harmonic generation (HHG) 41

42 dynamics and ultrafast spectroscopy in the attosecond time domain [6,7] as well as the sub-cycle

43 transient structures in time-dependent multiphoton ionization (MPI) processes [8,9]. These recent

44 studies have revealed novel transient multiphoton dynamics and spectroscopy in the ultrashort

45 time domain. In this paper, we focus on the exploration of the transient MPI dynamics and we

46 uncover the origin of the generation of the transient and distinct electron density structures seen

in the MPI processes for the first time by means of the Bohmian quantum trajectory approach.

48

In the study of strong-field HHG and MPI processes, the classical and semi-classical trajectory 49 methods have been widely used in the past and they are valuable in providing qualitative insight 50 51 regarding the multiphoton dynamics. For the HHG processes, the 3-step model [10,11] and strong-field approximation (SFA) [12] are often used. However, the SFA does not take into 52 account the Coulomb potential and electronic structure and cannot be used for the study of 53 54 below- and near- threshold multiphoton processes, for example. Fully ab initio quantum 55 mechanical solution of the time-dependent Schrödinger equation (TDSE) is currently feasible for one- and two- electron systems for the accurate treatment of MPI and HHG processes Bohmian 56 mechanics (BM) [13] is an alternative and complementary quantum approach which can provide 57 a trajectory-based scheme allowing for a causal interpretation of quantum mechanics. 58 Information on individual trajectories, along with the analysis of the emission times of various 59 groups of trajectories prepares a comprehensive and intuitive picture of the process under 60 investigation. Therefore, this method can serve in complement to the results obtained from direct 61 analysis of the electron density. The BM approach has been successfully applied to the model 62 study of problems such as photo-dissociation [14], tunneling [15], and atom diffraction by 63 64 surface [16], etc., in the past. More recently, it has been also used to the model study of strong field processes such as HHG [17], laser-driven electron dynamics [18-25], etc. 65

However, most of the BM studies of strong field processes so far have adopted either 1D or softpotential models. In this article, we present a fully *ab initio* 3D and accurate treatment of the
Bohmian trajectories beyond SFA, and discuss the formation of sub-cycle transient structures
seen in the MPI processes.

We treat the interaction of an intense laser field with a single hydrogen atom by solving the time-dependent Schrödinger equation (TDSE) (atomic units are used):

72

73

 $i\frac{\partial}{\partial t}\boldsymbol{\psi}(\boldsymbol{r},t) = \left[\hat{H}_{0} + \hat{V}(\boldsymbol{r},t)\right]\boldsymbol{\psi}(\boldsymbol{r},t), \qquad (1)$

74

where \hat{H}_0 is the unperturbed Hamiltonian of the hydrogen atom and $\hat{V}(r, t)$ is the time-dependent interaction of the electron with the laser field in the dipole approximation:

77

78
$$\hat{V}(\boldsymbol{r},t) = -\boldsymbol{F}(t)\boldsymbol{r} = -zF(t), \qquad (2)$$

F(t) being the force acting upon the electron from the laser field. For the sine-squared envelope 79 80 of the laser pulse.

$$F(t) = F_0 sin^2(\frac{\pi t}{T}) sin(\omega t), \qquad (3)$$

81 82

where F_0 is the peak field amplitude, ω is the carrier frequency, and T is a pulse duration. 83 Without loss of generality, we can assume that the polarization vector of the field lies in z-84 direction. In all our calculations we have used a laser pulse with the sine-squared envelope, total 85 duration of 20 optical cycles (o.c.), the carrier wavelength 800 nm (corresponding to the photon 86 energy 1.55 eV), and the peak intensity $8 \times 10^{13} W / cm^2$. 87

88

The time-dependent generalized pseudo-spectral (TDGPS) method [26] is used to solve the 89 TDSE in spherical coordinates accurately and efficiently. This method takes advantage of the 90 generalized pseudo-spectral (GPS) technique for non-uniform optimal spatial discretization of 91 the coordinates and the Hamiltonian using only a modest number of grid points. The time 92 propagation of the wave function under this method is performed by the split operator method in 93 94 the energy representation [26]: $\Psi(r, t + \Delta t)$

95

96
$$\cong \exp\left(-i\hat{H}_0\frac{\Delta t}{2}\right) \times \exp\left[-iV\left(\boldsymbol{r},\boldsymbol{\theta},t+\frac{\Delta t}{2}\right)\Delta t\right]$$

98

To impose correct outgoing-wave boundary conditions on the wave function and prevent 99 100 spurious

 $\times \exp\left(-i\hat{H}_{0}\frac{\Delta t}{2}\right)+O\left(\Delta t^{3}\right).$

reflections from the boundary of the spatial domain, we use an absorbing layer at large distances 101 core. The absorber implemented through 102 from the atomic is the mask function $\cos^{0.25}[\pi(r-r_0)/2(r_{\text{max}}-r_0)]$, $(r \ge r_0)$ with $r_{\text{max}} = 100a.u.$, $r_0 = 80a.u.$. The wave 103 function is multiplied by the mask function at each time step. Because of the absorber, the norm 104 of the wave function decreases in time. The time-dependent ionization rate can be defined as a 105 logarithmic derivative of the time-dependent population P(t) [8]: 106

107
$$P(t) = \int |\boldsymbol{\psi}(\boldsymbol{r}, t)|^2 d^3 r, \qquad (5)$$

108
$$\Gamma(t) = -\frac{d}{dt} ln P(t).$$
 (6)

109

(4)

For atoms in linearly polarized laser fields, the angular momentum projection onto the polarization direction of the field (the *z*-axis) is conserved. That means the dependence of the wave function on the angle φ (rotation angle about the *z*-axis) is reduced to the factor $\exp(im\varphi)$, where *m* is the angular momentum projection. For m=0 the wave function does not depend on φ at all, thus the gradient of the wave function ψ can be calculated with respect to the coordinates *r* (radial coordinate) and θ (angle between the radius-vector and *z*-axis):

$$\nabla \psi = \boldsymbol{e}_r \frac{\partial \psi}{\partial r} + \boldsymbol{e}_\theta \frac{1}{r} \frac{\partial \psi}{\partial \theta} = \boldsymbol{e}_r \frac{\partial \psi}{\partial r} - \boldsymbol{e}_\theta \frac{\sin\theta}{r} \frac{\partial \psi}{\partial \cos\theta} \,. \tag{7}$$

118

117

119 e_r and e_{θ} are the unit vectors of spherical coordinate system. The equation for the Bohmian 120 trajectories reads as

- 121
- 122

 $\frac{d\mathbf{r}}{dt} = Im \frac{\nabla \psi}{\psi} , \qquad (8)$

We note that our approach is fully based on quantum mechanics. The right-hand side of Eq.(8) represents the velocity field from an accurate quantum-mechanical wave function. Thus there is no discrepancy between the analysis based on quantum-mechanical fluxes and that based on the Bohmian trajectories. Since the velocity $\frac{d\mathbf{r}}{dt}$ has the following expansion in the spherical coordinate system,

127 coordinate system,

128

 $\frac{d\mathbf{r}}{dt} = \mathbf{e}_r \frac{dr}{dt} + \mathbf{e}_{\theta} r \frac{d\theta}{dt} + \mathbf{e}_{\varphi} r \sin\theta \frac{d\varphi}{dt},\tag{9}$

129

the vector equation (8) is equivalent to a set of three 1D equations:

131

132 $\frac{dr}{dt} = Im \left(\frac{1}{\psi} \frac{\partial \psi}{\partial r}\right), \tag{10}$

134
$$\frac{d\theta}{dt} = -\frac{\sin\theta}{r^2} Im \left(\frac{1}{\psi} \frac{\partial\psi}{\partial\cos\theta}\right), \tag{11}$$

135

 $\frac{d\varphi}{dt} = 0.$ (12)

136

Obviously, the angle φ does not change, and the trajectory lies in the plane defined by the initial (at $t = t_0$) radius-vector and the *z*-axis. One has to solve the Cauchy problem for the set of two equations (10) and (11). In the generalized pseudo-spectral (GPS) discretization, we use the Gauss-Lobatto scheme for the variable *r* (with the appropriate mapping transformation) and the 141 Gauss scheme for the variable $cos\theta$. The expression for the first derivative with respect to r142 appears as following:

143

144

$$\left(\frac{\partial\psi}{\partial r}\right)_{r(x_i)} = \frac{1}{r'(x_j)} \sum_{j'=1}^{N_x} \frac{P_{N_x+1}(x_j)}{P_{N_x+1}(x_{j'})} d_{jj'}^x \psi((x_{j'})), \qquad (13)$$

Here, P_{N_x+1} is the Legendre polynomial. N_x is the number of collocation points (roots of the derivative of Legendre polynomial, $P'_{N_x+1}(x)$), not including the end points -1 and +1. The matrix elements for the $d_{jj'}^x$ Gauss-Lobatto discretization are as listed below [27]:

148

149
$$d_{jj'}^{x} = \frac{1}{x_{j'} - x_{j}} (j \neq j'), \quad d_{jj}^{x} = 0 (j \neq 0, j \neq N)$$

 $d_{00}^{x} = -\frac{N(N+1)}{4} , \ d_{NN}^{x} = \frac{N(N+1)}{4},$ (14)

150 151

152 The first derivative with respect to $cos\theta$ is as follows:

153

154
$$\left(\frac{\partial\psi}{\partial\cos\theta}\right)_{\cos\theta_{j}} = \sum_{j'=1}^{N_{y}} \frac{P'_{N_{y}}\left(y_{j}\right)}{P'_{N_{y}}\left(y_{j'}\right)} d_{jj'}^{y} \psi\left(\left(\cos\theta_{j'}\right)\right)$$
(15)

155

Here N_y is the number of collocation points in the Gauss scheme (roots of the Legendre polynomial P_{N_y}). Eq.(15) assumes that the mapping transformation is just the identity transformation, i.e. $cos\theta = y$. The matrix elements d_{ij}^{y} are defined as following [28]:

159
$$d_{jj'}^{y} = \frac{1}{y_j - y_{j'}} \left(j \neq j' \right), \quad d_{jj}^{y} = \frac{y_j}{1 - y_j^2}, \tag{16}$$

160

161 To calculate the first derivative of the Legendre polynomials, one can use the following 162 recursion relation:

163
$$P_{N_{y}}^{'}(y) = \frac{N_{y}(N_{y}+1)}{(2N_{y}+1)(1-y^{2})} \Big[P_{N_{y}-1}(y) - P_{N_{y}+1}(y) \Big].$$
(17)

164

The set of coupled ordinary differential equations (10) and (11) is solved numerically with the help of the 4th order Runge-Kutta (RK4) method, yielding the electron quantum trajectories. Since the quadrature points for RK4 differ from the original GPS grid points, we need to perform an additional interpolation using the GPS interpolation formula [29], to be able to evaluate the numerical values of the wave function at the coordinate points supplied by the RK4 solver:

171
$$\psi(r,\theta,t) = \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} \psi(r_i,\theta_j,t) \frac{P_{N_x+2}(x) - P_{N_x}(x)}{(2N_x+3)(x-x_i)P_{N_x+1}(x_i)} \times$$

172
$$\frac{(2N_{y}+1)(1-y_{j}^{2})P_{N_{y}}(y)}{N_{y}(N_{y}+1)(y-y_{j})[P_{N_{y}-1}(y_{j})-P_{N_{y}+1}(y_{j})]}.$$
 (18)

In Fig.1, we use Bohmian trajectories to illustrate the dynamics of electron and its ionization rate within 7 optical cycles (5th-11th) of the given laser field, Fig.1(a). From left to right in Fig.1(b) one can see how distinct groups of Bohmian trajectories eventually are being formed at higher optical cycles. During the 5th optical cycle (the left most panel, shaded in green in Fig.1(b)) only one group of trajectories is distinguishable. As will be discussed later, these trajectories are representing a single portion of electron density detached from the core toward negative zdirection. When the laser electric field changes sign, some portion of these trajectories change direction and travel back to the core. The retuning process happens in a time interval of about 0.5 o.c. Since the returning trajectories travel different distances before they change direction and return to the core, each of them would have different return energies. This causes transitions to excited bound and continuum states of the unperturbed atom over time, resulting in the oscillations of the electron density. Therefore when the next ionization is about to happen toward negative z-direction, these oscillations of the electron density give rise to multiple wave packets, instead of just one. As can be seen in Fig.1(b) this effect becomes more and more influential at higher optical cycles up to the maximum laser intensity (11th optical cycle). By symmetry, the similar behavior happens for the trajectories travelling toward positive z-direction within the second half of each optical cycle. This effect is clearly pronounced as multiple bursts in the MPI rate diagram, Fig.1(c). We will discuss this in more detail later on in this article.



200

FIG.1. (Color online) (a) The 800 nm sin^2 driving laser pulse with the peak intensity of 8×10¹³*W*/*cm*². Shaded in different colors, are the regions where Bohmian trajectory calculations are performed. (b) Bohmian trajectories computed within the time intervals given in (a). (c) Corresponding ionization rate plots within first-half optical cycle of the investigated time intervals. Distinct groups of electron trajectories eventually form from left to right, which in turn cause multiple bursts in ionization rate.

A detailed picture of the electron dynamics during the ionization process can be achieved by combining 207 an analysis of the Bohmian trajectories and that of the time evolution of the electron density. In Fig.2, 208 the evolution of the electron density of hydrogen atom is presented within the 11th o.c. of the 209 laser field. As illustrated in this figure, several distinct density portions are shaped and detached 210 from the atom within a half optical cycle of the laser field. These multiple detachments of 211 electron density from the core happen at different times, not necessarily when the external field 212 reaches its maximum value. The oscillations of the electron density are caused by transitions to 213 excited bound and continuum states in the laser field [8]. Besides the portions of electron density 214 that leave the core after direct ionization, some detached wave packets return to the parent ion 215 when the force from the laser field becomes positive (after 0.25 o.c.). As illustrated by the red 216 and green colors, some of these wave packets travel longer (L), and some have shorter travel 217 time (S), respectively, before they can return to the core. Bohmian trajectories corresponding to 218 each of these wave packets will be further analyzed below. 219





221







FIG.2. (Color online) Sub-cycle electron density evolution of the hydrogen atom in the 800 nm sin^2 laser pulse with the peak intensity of $8 \times 10^{13} W / cm^2$. Horizontal axis is the distance in the z-direction and the vertical axis shows the distance in the x-direction. The laser is polarized in the z-direction. The inset shows the corresponding phase of the laser field. The direction of the force from the laser field is illustrated by a small arrow. The laser field in the inset is shown for the interval -0.5 to +1.0 o.c. with the grid lines spacing of 0.1 o.c. The red and green regions represent the wavepackts returning to the core after a longer (L) and shorter (S) travel time, respectively. The color scale for the density is logarithmic.

- .0

In order to explore the dynamical mechanism of electron wave packet motion, we have performed semi-classical calculations, extending the standard approach suggested independently by Corkum [10] and Kulander et al. [11], with the inclusion of the Coulomb potential. Here, the electric force corresponding to the applied laser field is $F_z = F(t)e_z$, where e_z is the unit vector in the z-direction and F(t) is given by Eq. (3). When F_z and the electron velocity have the same direction, the electron gains the energy to escape the potential well. In Fig. 3(a), the result of the semi-classical approach is presented as electron return energy versus time. The initial conditions are set as $x_0 = y_0 = 17$ a.u, $z_0 = 0$, $v_{x0} = v_{y0} = 0$, $v_{z0} = -0.1$ a.u. The return energy is calculated as $E_k + E_p$, where E_k and E_p are the kinetic and Coulomb potential energies, respectively, when z coordinate of the electron returns to its initial value z = 0.0a.u. Here, we can indicate the short and long trajectories as those in the standard three-step model. As shown in this figure, the trajectories that are released late and return early are regarded as the short trajectories (green region), while those released early and returned late are the long trajectories (red region). The corresponding time intervals are shaded with the same colors in Fig. 3(b) on the laser field plot. Here the labels b through j correspond to the time moments for which the electron densities are presented in Fig. 2. The important feature of this result is the agreement between the ionization and return time periods predicted by semi-classical results and the one obtained from analyzing the electron density evolution and also determined by Bohmian trajectory calculations. The later result will be discussed in detail next.



FIG.3. (Color online) (a) Semi-classical return energy as a function of the return time. The red and green lines indicate the long and short trajectories, respectively. (b) Driving laser pulse. Red and green shaded areas correspond to the long and short electron return times. The red and green dots represent samples for each set of trajectories. The curved red and green arrows illustrate, schematically, the long and short and long trajectories. The labels b through j correspond to the time moments for which the electron densities are presented in Fig.2.

Fig. 4(a) shows the wave packets detached from the hydrogen atom at 0.19 o.c. (with respect to the beginning of the 11th o.c.) with different colors. In general, there is a close resemblance of the trajectory patterns in adjacent optical cycles except at the beginning and the end of the laser pulse where the intensity is very low. In Fig. 4(b), we report the corresponding groups of the Bohmian trajectory flows with the same color and labels. To generate the results presented in Fig. 4(b), the initial time for RK4 solver in the current case is set to -0.25 o.c. The initial position of the electron is scanned between $z_0 = 1$ and $z_0 = 10$ a.u., with $x_0 = 1$ a.u. Within each half optical cycle, the outermost wave packets continue moving towards larger distances from the core, thus describing ionization. The corresponding trajectories belonging to this group are indicated by the label "5". Please note that the ionization probability at the end of the laser pulse, calculated according to Eq. (5), is about 1.4 percent only. The wave packets (and the corresponding trajectories) labeled by "4" and "3" are the ones that oscillate in the external field before leaving the area shown in Fig. 4, without returning close to the core. Labels "2" and "1" indicate some longer and shorter trajectories, respectively. These trajectories represent the travelling wave packets in Fig. 2, which are shaded with the same colors. As can be seen in Fig. 4(b), the longer trajectory "2" reaches its maximum distance (around z = -30 a.u.) at about 0.25 o.c. After this point it travels back and returns to the core at about 0.68 o.c. The shorter (green) trajectory "1", on the other hand, travels in z-direction to about -15 a.u and then returns back to the target at about 0.37 o.c. (see Fig. 2(h)). This group of trajectories represents the motion of the green shaded wave packet in Fig. 2(d-h). In Fig. 4(b), one can also see the trajectories featuring two returns to the parent ion (blue line). Finally, the innermost trajectories (intense gray area below z = -5.0 a.u.) have smaller momenta and are mainly governed by the Coulomb potential. These trajectories vibrate in a short distance range around the nucleus and describe the dynamical aspects of lower bound states. These results are in complete agreement with the semi-classical picture presented in Fig. 3(a).



327

FIG.4. (Color online) (a) Multiple portions of the electron density detached from the hydrogen 328 atom as seen at 0.19 o.c. from the beginning of the 11th o.c. in the sin² laser pulse with the 329 carrier wavelength 800nm. Different colors indicate different portions of the electron density 330 (also labeled by "1" to "5"). (b) A thousand of the Bohmian trajectories initiated at 331 $t_0 = -0.25$ o.c. representing the time evolution of the hydrogen atom in the same laser field. The 332 labels "1" and "2" indicate the groups of short and long trajectories, respectively. One trajectory 333 in each group is represented by a bold line. The groups of trajectories labeled by "3" and "4" 334 represent the wave packets that oscillate in the external field without revisiting the core. The 335 trajectories "5" represent the motion of the outermost wave packet. The blue single line shows 336 the trajectory that returns twice to the nucleus. The intense gray area under z = -5.0 a.u.337 corresponds to short oscillatory trajectories close to nucleus. 338

Recently, Telnov *et. al.* reported multiple ionization bursts within a single optical cycle in the 339 time-dependent ionization rate of the hydrogen atom [8]. The ionization rate is defined by Eqs. 340 (5) and (6) for the whole spherical volume with the radius r_{max} where the time-dependent 341 Schrödinger equation is solved. However, we can also define this rate on the boundary of a 342 smaller spherical volume with the radius $r_c < r_{max}$. Defined in this way, the quantity dP/dt343 represents the electron current through the sphere of radius r_c . Certainly, this depends on both 344 the time and radius r_c . The time profile of the rate dP/dt changes with the distance r_c [8]. Here 345 we report the sub-cycle structures in the time-dependent rates calculated on the distances as 346 small as few tens of atomic units from the core. We understand that direct measurements of the 347 electron current close to the atomic core could be extremely difficult or even implausible. 348 Experimental observations are more feasible at sufficiently large distances from the target. Of 349 course, in the far asymptotic region, the time-dependent signal would be reshaped due to 350 different times of flight of the electrons with different energies. Still, the information about the 351 sub-cycle structures of the electron current at smaller distances must be encoded in that signal. 352 Then a theoretical procedure can be applied that maps the properties of the outgoing-wave packet 353 at large distances to earlier times and smaller 350 distances. Construction of such a procedure 354 can be a subject of a separate study. In Fig. 5(a) we present the time-dependent ionization rate for 355 the few central optical cycles for $r_c = 25 a.u$. One can find in this figure a similar pattern of four 356 bursts at each half optical cycle. The structure of the ionization rate can be explained using the 357 Bohmian trajectories studied here, as well as the electron densities (Fig.2). As it was discussed in 358 [8], the portions of the outgoing wave packet, created under the influence of the external field, 359 contain states belonging to various energies. Different groups of electrons travelling along 360 distinct trajectories can represent this various energy contributions. The Bohmian trajectories in 361 the 11th o.c. optical cycle are shown in Fig. 5(b). As it was mentioned above, the same pattern is 362 observed in other time intervals and the reason for chosing this optical cycle is that the laser 363 intensity is around its maximum. In Fig. 5(b) four groups of trajectories are shaded with different 364 colors (labeled A to D). These groups are responsible for the bursts observed in the ionization 365 rate. Apparently, the intensity of each peak in the ionization rate is related to the number of the 366 Bohmian trajectories found in the corresponding group. Peak A corresponds to the trajectories 367 that represent the first (small) wave packet that does not came back to the core. The next peak 368 (B) corresponds to a more distributed wave packet, which oscillates at a large distance from the 369 core before it eventually leaves the parent ion. The next group of trajectories represents the wave 370 371 packets created at smaller distances from the nucleus. As one can see in Fig. 4(b), the trajectories C and D can come back closer to the core before they go to large distances describing ionization. 372 Shorter (green) trajectories never reach to -25 a.u., and therefore do not contribute to the 373 ionization rate. Analysis of the Bohmian trajectories, as one can see, provides a simple and 374 intuitive explanation of such a subtle phenomenon as multiple bursts in the time-dependent 375 ionization rate. 376



FIG.5. (Color online) (a) Time-dependent ionization rate of the hydrogen atom within a half optical cycle. (b) Four distinct groups of the Bohmian trajectories (labeled A through D), within a half optical cycle, shaded with different colors corresponding to the peaks in the timedependent ionization rate. The red dashed line shows $r_c = 25a.u$.

In summary, we presented a fully *ab initio* 3D and accurate treatment of the Bohmian trajectories 384 beyond SFA, and used it to illustrate the formation of sub-cycle transient structures seen in the 385 MPI processes. Electron wave packet dynamics and ionization process on a sub-femtosecond 386 time scale for the hydrogen atom subject to intense near-infrared laser fields were analyzed. As a 387 388 complementary tool, an accurate treatment of the electron dynamics in the De Broglie-Bohm's framework of the Bohmian mechanics was introduced to investigate the multiple peaks structure 389 of the time-dependent ionization rate within a half optical cycle. The nature of this phenomenon 390 was revealed by the analysis of the time-dependent electron density and the Bohmian trajectories 391 representing different groups of electrons with various ionization times and pathways. The 392 external field causes transitions to the excited bound and continuum states of the unperturbed atom in the 393 394 course of time, resulting in the oscillations of the electron density. A nonlinear response of the electron density to the laser field through transitions to the excited states leads to shaping of multiple density 395 portions detached from the core during each optical cycle. In the Bohmian trajectory analysis, it is 396 397 reflected in formation of distinct groups of the trajectories describing ionization. This is the origin of multiple ionization bursts per optical cycle. We have also performed semi-classical simulations, which 398 illustrate various energy contributions to the wave packet created under the influence of the external laser 399 400 field.

401

This work was partially supported by the Chemical Sciences, Geosciences and Biosciences 402 Division of the Office of Basic Energy Sciences, Office of Sciences, U.S. Department of Energy. 403 We also acknowledge partial support from the Ministry of Science and Technology of 404 Taiwan and National Taiwan University (Grants No. 104R104021 and No. ERP-104R8700-2). 405 406 D.A.T. acknowledges partial support from St. Petersburg State University (Grant No. 11.38.654.2013). P.C.L. acknowledges partial support from National Natural Science 407 Foundation of China (Grants No. 11364039 and No. 11465016), Natural Science Foundation of 408 Gansu Province (Grant No. 1308RJZA195), and Education Department of Gansu Province 409 (Grant No. 2014A-010). 410

411

412 **References**

- 413
- 414 [1] F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009).
- 415 [2] P. B. Corkum and F. Krausz, Nature Phys. **3**, 381 (2007).
- 416 [3] S. Chen, M. J. Bell, A. R. Beck, H. Mashiko, M. Wu, A. N. Pfeiffer, M. B. Gaarde, D. M. Neumark,
 417 S. R. Leone, and K. J. Schafer, Phys. Rev. A 86, 063408 (2012).
- 418 [4] M. J. Bell, A. R. Beck, H. Mashiko, D. M. Neumark, and S. R. Leone, J. Mod. Opt. 60, 1506 (2013)
- 419 [5] M. Chini, X. Wang, Y. Cheng, Y. Wu, D. Zhao, D. A. Telnov, S. I. Chu, and Z. Chang, Sci. Rep. 3,
 420 1105 (2013).
- 421 [6] J. Heslar, D. A. Telnov, and S. I. Chu, Phys. Rev. A 89, 052517 (2014).
- 422 [7] K. N. Avanaki, D. A. Telnov, and S.I Chu, Phys. Rev. A 90, 033425 (2014).
- 423 [8] D.A. Telnov, K.N. Avanaki, and S.I. Chu, Phys. Rev. A 90, 043404 (2014).
- 424 [9] N. Takemoto and A. Becker, Phys. Rev. Lett. 105, 203004 (2010).

- 425 [10] P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).
- 426 [11] K. C. Kulander, K. J. Schafer, and J. L. Krause, Proceedings of the Workshop on Super-
- Intense Laser Atom Physics (SILAP) III, edited by P. Piraux (Plenum Press, New York) 316,
 95(1993).
- 429 [12] M. Lewenstein, P. Salieres, and A. L'Huillier, Phys. Rev. A 52, 4747 (1995).
- 430 [13] D. Bohm, Phys. Rev. 85, 166 (1952).
- 431 [14] F. S. Mayor, A. Askar, and H. Rabitz, J. Chem. Phys. 111, 2423 (1999).
- 432 [15] C. L. Lopreore and R. E. Wyatt, Phys. Rev. Lett. 82, 5190 (1999).
- 433 [16] R. Guantes, A. Sanz, J. Margalef-Roig, and S. Miret-Artes, Surf. Sci. Rep. 53, 199 (2004).
- 434 [17] J. Wu, B. B. Augstein, and C. F. d. M. Faria, Phys. Rev. A 88, 023415 (2013).
- 435 [18] P. Botheron and B. Pons, Phys. Rev. A 82, 021404 (R) (2010).
- 436 [19] R. Sawada, T. Sato, and K. L. Ishikawa, Phys. Rev. A 90, 023404 (2014).
- 437 [20] J. Wu, B. B. Augstein, and C. F. de Morisson Faria, Phys. Rev. A 88, 063416 (2013).
- 438 [21] S. S. Wei, S. Y. Li, F. M. Guo, Y. J. Yang, and B. Wang, Phys. Rev. A 87, 063418 (2013).
- 439 [22] J. Stenson, and A. Stetz, Eur. J. Phys. **34**, 1199 (2013).
- 440 [23] N. Takemoto, and A. Becker, J. Chem. Phys. 134, 074309 (2011).
- 441 [24] S. Dey, and A. Fring, Phys. Rev. A 88, 022116 (2013).
- 442 [25] Y. Song, F. M. Guo, S. Y. Li, J. G. Chen, S. L. Zeng, and Y. J. Yang, Phys. Rev. A 86,
- 443 033424 (2012).
- 444 [26] X. M. Tong, and S. I. Chu, Chem. Phys. 217, 119 (1997).
- 445 [27] D. A. Telnov and S. I. Chu, Phys. Rev. A 59, 2864 (1999).
- 446 [28] D. A. Telnov and S. I. Chu, Phys. Rev. A 76, 043412 (2007).
- 447 [29] D. A. Telnov and S. I. Chu, Phys. Rev. A 79, 043421 (2009).