

Helium atom in intense and superintense laser fields: A new theoretical approach

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Abstract. A quantum hydrodynamical study is made of the dynamical changes of a helium atom interacting with lasers of two different intensities, but having the same frequency. Under the intense laser field, electron density oozes out of the helium atom by absorbing laser photons and getting promoted to higher excited states including the continuum. Under the superintense field, electron density partly moves away from the helium nucleus but remains in the “quasi-bound” dressed states along with the laser field, thus suppressing ionization.

Keywords. Helium; laser fields.

Laser pulses are known to control a particular atomic or molecular process. For example, one bond in a polyatomic molecule can be broken selectively [1] by properly designing the laser field. Similarly, an atom can exhibit different types of behaviour when probed by a laser of controlled intensity, e.g., atoms can undergo ionization (three possible types of ionization are: above-threshold [2, 3], tunneling [4] and sequential [5, 6]), excitation leading to emission of radiation [7, 8] (including high-order harmonic generation) and stabilization with respect to ionization [9, 10]. Here, we report a new theoretical, quantum hydrodynamical study of the dynamical changes of a helium atom probed by lasers of two different intensities (I) having the same frequency (0.1382 a.u.). When $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ (intense field) electron density oozes out (ionization) of the helium atom by absorbing laser photons and getting promoted to higher excited states including the continuum. However, when $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ (superintense field), electron density partly moves away from the helium nucleus but remains in the “quasi-bound” dressed states along with the laser field, thereby suppressing ionization. We provide support for this conclusion by calculating the extra stabilization energy gained by the electrons in the presence of the laser field. The non-perturbative methodology devised in our laboratory is applicable to any time-dependent quantum mechanical process.



We consider the electron density in an atom as that of a continuous “classical” fluid which obeys two quantum mechanical equations [11, 12] viz., a continuity equation and a Navier–Stokes-type equation of motion, as follows:

$$\partial_t a^2 + \nabla_q \left(a^2 \frac{\nabla_q S}{m} \right) = 0; \quad \partial_t = \frac{\partial}{\partial t}, \quad (1)$$

$$\partial_t S + \frac{(\nabla_q S)^2}{m} - \frac{\hbar^2 \nabla_q a}{2m a} + V_c^{qu}(a) + V(q, a) = 0, \quad (2)$$

where $V_c^{qu}(a)$ is a non-classical additional term needed for more than two-electron systems, which gives the additional kinetic energy in addition to the Weizsäcker contribution. $V(q, a)$ is a density (a^2)-dependent non-linear potential given by

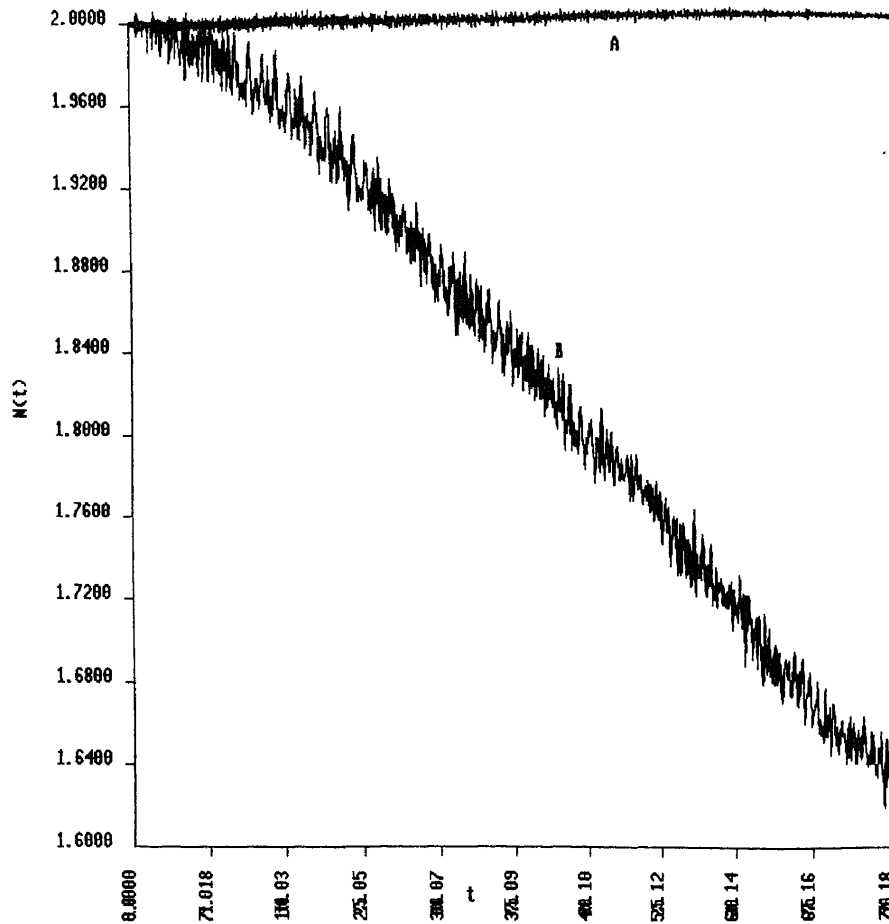


Figure 1. Calculated total electronic charge (a.u.) at different times. A decrease of total charge from initial ($t = 0$) value of 2 to a final ($t = 750.18$ a.u.) value of 1.63 is noticed for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ (B) whereas for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ (A) total charge remains practically fixed at 2.

He atom in intense and superintense laser fields

(x, exchange; c, correlation; I, interaction)

$$V(q, a) = \frac{\delta E_x[a]}{\delta a} + \frac{\delta E_c[a]}{\delta a} + \frac{\delta E_{el-el}[a]}{\delta a} + \frac{\delta E_{nu-el}[a]}{\delta a} + \frac{\delta E_I[a]}{\delta a}. \quad (3)$$

Under the construction of an effective hamiltonian, H_{eff} is given as

$$H_{eff} = -\frac{\hbar^2}{2m} \nabla_q^2 + V(q, a) + V_c^{qu}(a), \quad (4)$$

and a hydrodynamical "wave" function,

$$\Psi_{hy} = ae^{iS/\hbar}; \quad a, S \text{ real} \quad (5)$$

which gives a collective description of all the electrons, we obtain a generalized non-linear Schrödinger equation (GNLSE) in three-dimensional space unlike the conventional Schrödinger equation in 3N-dimensional configuration space. The GNLSE may be written as

$$i\hbar \frac{\partial \Psi_{hy}(q)}{\partial t} = H_{eff} \Psi_{hy}(q). \quad (6)$$

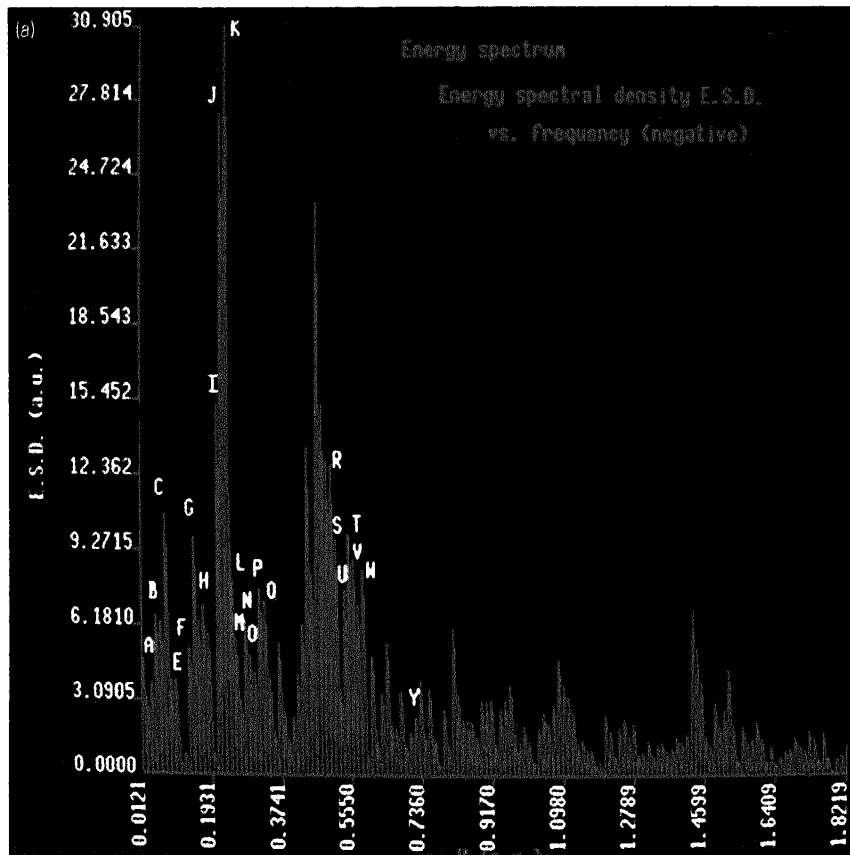


Figure 2a.

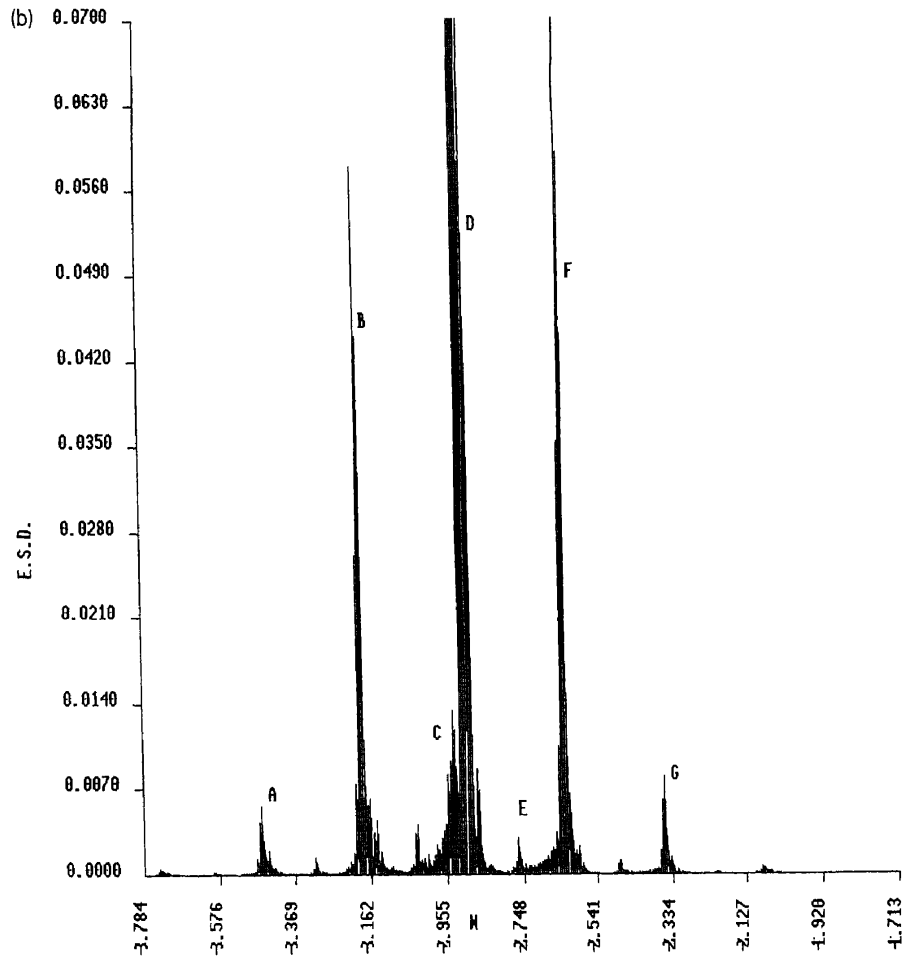


Figure 2. Calculated energy spectrum (a.u.) showing the spectral density (E.S.D.) for different states of energy, W . A large number of states with different labels signifying higher excitation and consequent ionization can be seen for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ (a). For $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ (b), only a few quasi-bound “dressed” states are noticed. Some of the quasi-bound states have lower energy than the ground state energy of He atom (b).

This formulation is closer to the classical description of quantum systems in the sense that the reduction of $3N$ degrees of freedom in the Schrödinger wave function to 3 in the hydrodynamical wave function (eq. (5)), results in the loss of interference—a characteristic feature of quantum systems. Thus, Ψ_{hy} is essentially a “reduced” form of Schrödinger wave function.

To understand the behavioural change of atomic systems under varying laser fields, one often resorts to varying techniques ranging from time-dependent Schrödinger equation and time-dependent Hartree–Fock equation to Kramer–Henneberger-frame classical-type calculation. We report some of the results obtained through the above

He atom in intense and superintense laser fields

quantum hydrodynamical methodology for two laser fields, (i) intense field with $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ and (ii) superintense field with $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$, both having the same frequency. For the He atom, we perform numerical evaluation of the hydrodynamical 'wave function' in the discrete cylindrical space (ρ, \tilde{z}) over a large number of time steps, each of which amounts to propagating Ψ_{hy} forward by an amount Δt , given by

$$\Delta t = \begin{cases} \frac{\pi}{2700 \omega_L}, & \text{up to the 30th time-step} \\ \frac{\pi}{90 \omega_L}, & \text{after the 30th time-step} \end{cases} \quad (7)$$

The above propagation is done with a coherent oscillating laser pulse of frequency, $\omega_L = 0.1382$ a.u. propagating along the axis (\tilde{z}) of the cylinder given by

$$E(t) = E_0 f(t) \sin(\omega_L t), \quad (8)$$

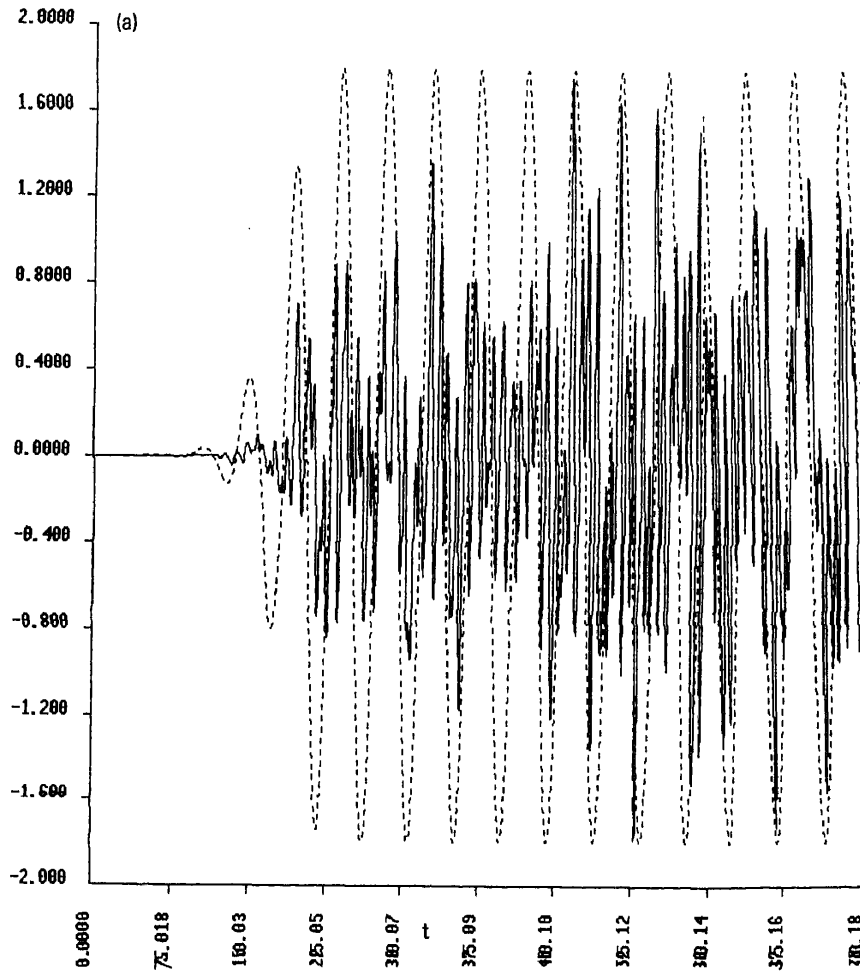


Figure 3a.

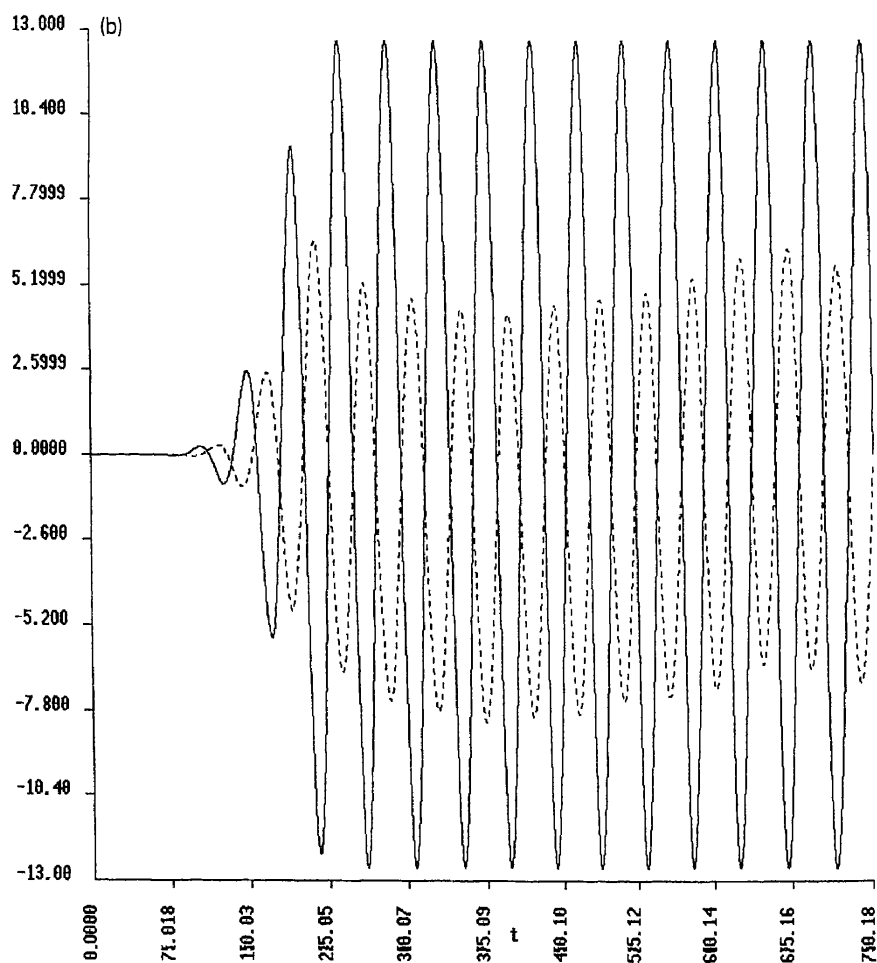


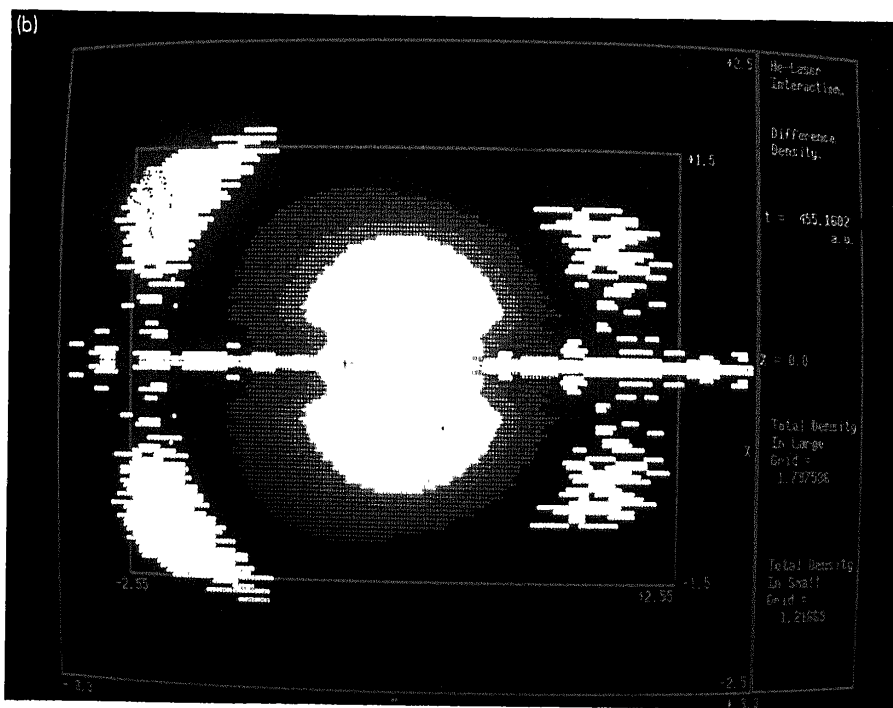
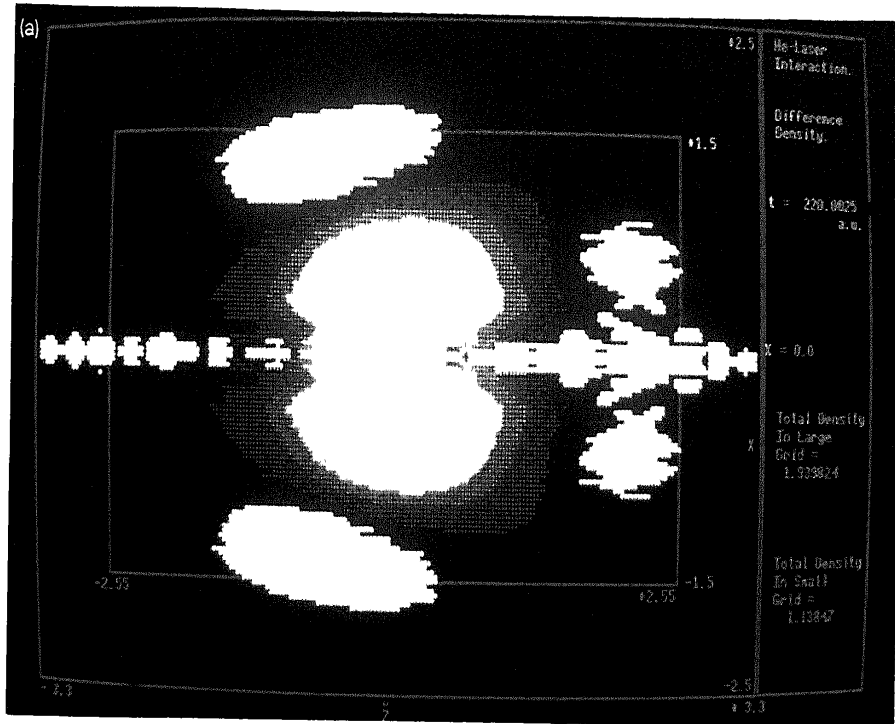
Figure 3. Average \bar{z} -directional expectation value (a.u.) of electronic charge distribution (solid line) along with the laser electric field (dotted line) for both the lasers. The two oscillations are seen to be non-resonant for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ (a) and the amplitude for electronic charge oscillation (solid line) is much higher than that for electric field (dotted line). In figure 3a, the ordinate is to be divided by 45 for the electric field (dotted line). For $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ (b), the two oscillations are seen to be resonant with little phase difference. Here the amplitude of electronic charge oscillation (dotted line) is much lower than that of electric field (solid line). In figure 3b, the ordinate is to be divided by 15 for the electronic charge oscillation (dotted line).

where the pulse-shape function, $f(t)$ is given by

$$f(t) = \begin{cases} e^{-\alpha(t-t_0)^2}, & t < t_0 \\ 1, & t \geq t_0. \end{cases} \quad (9)$$

Starting from the ground-state Hartree-Fock density of the He atom, the results of our numerical solution are displayed in figures 1 to 4. In figure 1, we show the explicit

He atom in intense and superintense laser fields



Figures 4a, b.

ionization of He atom (decrease in the electronic charge, N) under $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ whereas no ionization ($N = 2$ remains practically fixed) is seen for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$. The ionization at $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ is due to the higher absorption of energy out of the laser photons, leading to a dynamical state having contributions from higher singly-excited, doubly-excited (autoionizing) and continuum states of He atom. The presence of a large number of these states can be seen from the energy spectrum displayed in figure 2a. The same plot (figure 2b) for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$, however, shows only a few low-lying quasi-bound "dressed" states, some of whose energies lie lower than the ground-state energy of He atom (-2.90312 a.u.). Thus, very little absorption leading to excitation occurs for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$. At first sight this might appear to be counter-intuitive, but the qualitative explanation of this phenomenon is relatively simple.

The high absorption of laser photons for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ is attributed to the fact that the oscillations of electronic charge density along the polarization direction of the laser field cannot follow those of the laser field and thus the two types of oscillations show a pronounced phase-difference—a non-resonant phenomenon. However, for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$, the atom-laser interaction is nearly resonant with little phase difference (maximum, 1.13 rad.) between the two oscillations. This can be seen from figures 3a and 3b which display the z -directional expectation value of the electron density and electric field for both the lasers. The small phase difference at $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ is the cause of the small absorption, excitation and practically no ionization. In other words, at the superintense field, electronic behaviour is dictated by the laser field rather than the nuclear field while at the intense field, the nuclear field still dominates over the laser field.

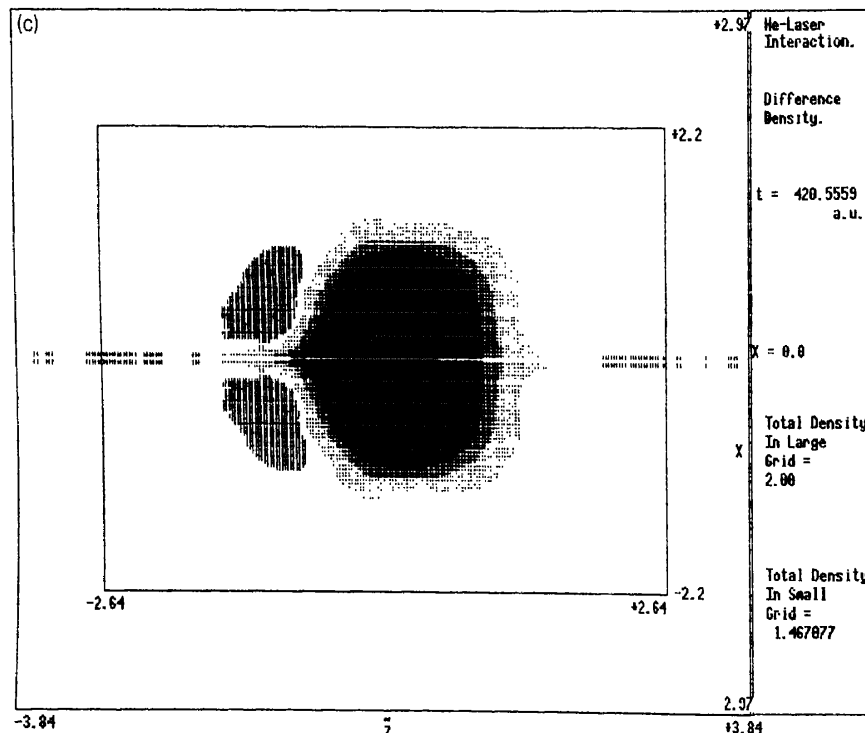


Figure 4c.

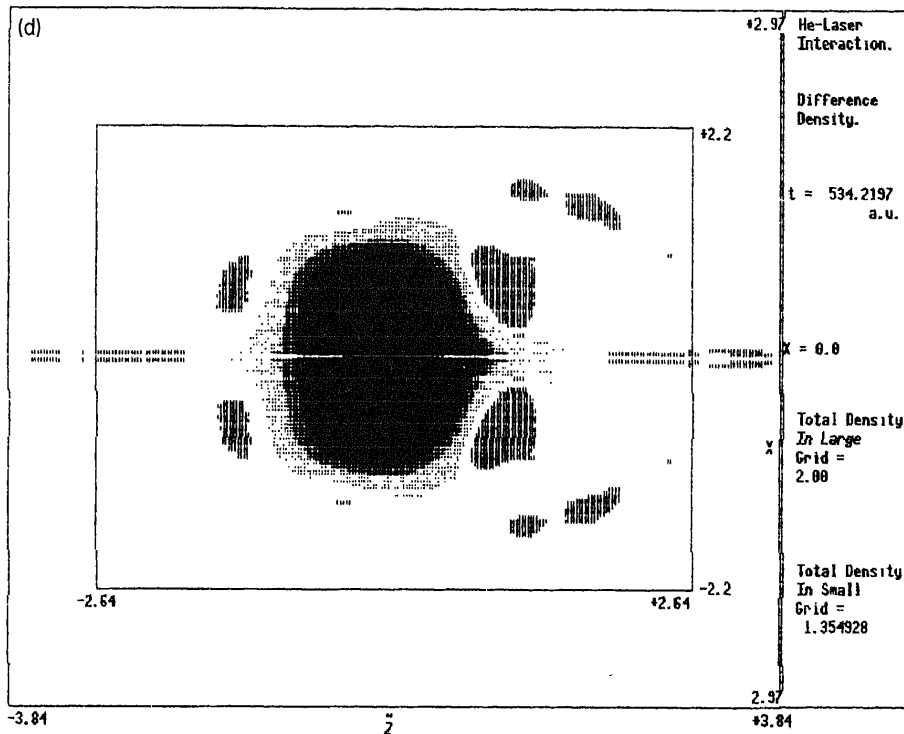


Figure 4. Difference electronic charge density (a.u.) in the $(\tilde{\rho}, \tilde{z})$ plane for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ at $t = 220.0025 \text{ a.u.}$ (a), $t = 455.1602 \text{ a.u.}$ (b) and for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ at $t = 420.5559 \text{ a.u.}$ (c), $t = 534.2197 \text{ a.u.}$ (d). A small window inscribed into the large grid monitors the departure of electronic density from the nuclear site. In figures 4a and 4b, the magenta-coloured region signifies an increase in the density and the blue-coloured onion-layered region signifies a decrease of the same whereas in figures 4c and 4d the increase and decrease in the density are denoted by dots and lines, respectively.

For $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$, the large number of photons provides the electrons an extra stabilization energy, $E_{\text{stab}} = -\int \tilde{z} E(t) \rho(\mathbf{r}) d\mathbf{r}$ ($E_{\text{stab}} = -6.013 \text{ a.u.}$ at $t = 602.165 \text{ a.u.}$) which is higher than that of the electron-nuclear attraction energy ($E_{\text{nu-el}} = -3.659 \text{ a.u.}$ at $t = 602.165 \text{ a.u.}$) whereas for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ the extra stabilization energy ($E_{\text{stab}} = -0.046 \text{ a.u.}$ at $t = 602.165 \text{ a.u.}$) is very small compared to electron-nuclear attraction ($E_{\text{nu-el}} = -2.1488 \text{ a.u.}$ at $t = 602.165 \text{ a.u.}$). This difference in the two E_{stab} values is responsible for higher excitation/ionization at $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$ and extra stabilization for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$.

The significant difference in the behaviour of electron density for the two lasers in the $(\tilde{\rho}, \tilde{z})$ -space can be vividly seen from the plots (figure 4) of difference electronic density, $\Delta\rho = \rho(\mathbf{r}, t) - \rho(\mathbf{r}, 0)$ showing the topography of positive and negative values of difference density. These plots also show that electronic movement follows the electric field more closely for $I = 5.6 \times 10^{18} \text{ W cm}^{-2}$ than for $I = 5.6 \times 10^{13} \text{ W cm}^{-2}$. A decrease in the total density, from both small and large grids is seen in

figures 4a and 4b whereas figures 4c and 4d show a fall in density in the small grid but fixed in the large grid.

The above suppression of ionization for superintense laser field will sustain till the laser field is present, provided no nonlinear dissipative mechanism leading to the loss of photons is introduced during the interaction.

Our primary goal in this work has been to demonstrate that time-dependent quantum fluid density functional theory can be applied in a unified manner to explain the detailed features in the dynamical evolution of atoms under various time-dependent processes occurring generally in femtoseconds. Manifold aspects of these phenomena will be published elsewhere (see also ref. [6]).

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