

Validity of Time-Dependent Hartree-Fock Theory for the Multiphoton Ionization of Atoms

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The validity of time-dependent Hartree-Fock (TDHF) theory is explored for the multiphoton ionization of atoms. The 3D electrostatic interaction is replaced by a 1D soft-core interaction which allows the construction of an N -electron atom in only N dimensions. For multiphoton ionization of a two-electron "He" atom in its ground state, we solve the time-dependent Schrödinger equation directly on a 2D lattice and solve a 1D formulation of the TDHF equations. The 2D lattice solution compares favorably with the frozen-core TDHF results, but not with the full nonlinear TDHF results.

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Because of advances in high-intensity laser systems, experiments can now be performed in which the strength of the time-varying electromagnetic forces equals or surpasses the electron binding force of an atom. These types of experiments¹⁻³ have stimulated the development of various theoretical approaches⁴⁻¹³ to the approximate solution of Schrödinger's equation for this essentially nonperturbative regime of atomic physics. The direct solution of the time-dependent Hartree-Fock (TDHF) equations, which has been used for many years to study heavy-ion collisions in nuclear physics,¹⁴⁻¹⁷ has been recently applied to calculate multiphoton ionization rates in rare-gas atoms.^{18,19} In this Letter the validity of TDHF theory for the multiphoton ionization of closed-shell atoms is explored within the context of a simple model. By comparing an "exact" time-dependent solution of the model with the approximate TDHF results, we find that caution must be exercised when using the full nonlinear TDHF method to describe the multiphoton ionization of an atom. Our conclusions are in keeping with recent studies of multiphoton ionization of helium,

in which the full nonlinear TDHF method led to unphysical and ambiguous ionization rates,¹⁸ and recent studies of prompt nucleon emission from heavy-ion collisions, in which frozen-core TDHF solutions are modified in a perturbative manner to handle nucleon correlation effects.²⁰⁻²²

Many of the qualitative features of atoms in intense fields (such as above-threshold ionization, harmonic generation of light, and stabilization against ionization) have been successfully modeled^{9,23,24} using a 1D soft-core potential of the form $V(x) = -(1+x^2)^{-1/2}$. The bound eigenfunctions alternate parity and their eigenenergies follow Rydberg scaling. The idea may be extended to model an N -electron atom in N dimensions by replacing the 3D electrostatic interaction by a 1D soft-core interaction,

$$\frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} \rightarrow \frac{1}{(c_p + x_i^2 + x_j^2 + 2c_r|x_i||x_j|)^{1/2}}, \quad (1)$$

where c_p and c_r are arbitrary coefficients and atomic units are used. The Hamiltonian for the N -electron "atom" may thus be written as

$$H_{\text{atom}} = \sum_{i=1}^N \left[-\frac{1}{2} \frac{d^2}{dx_i^2} - \frac{z}{(c_p + x_i^2)^{1/2}} \right] + \sum_{i>j} \frac{1}{(c_p + x_i^2 + x_j^2 + 2c_r|x_i||x_j|)^{1/2}}, \quad (2)$$

where a nucleus of charge Z is located at $x=0$. The time-dependent Schrödinger equation is given by

$$\frac{i \partial \psi(\mathbf{x}, t)}{\partial t} = \left[H_{\text{atom}} - iW(\mathbf{x}) + E(t) \sin(\omega t) \sum_{i=1}^N x_i \right] \psi(\mathbf{x}, t), \quad (3)$$

where $W(\mathbf{x})$ is an absorbing potential at the boundary, and $E(t)$ is the amplitude and ω is the frequency of the electromagnetic field.

The time-dependent Schrödinger equation (3) may be solved directly for the ground state of a two-electron "He" atom by application of the variational principle on a large 2D lattice. The resulting finite-difference equations for grid

points $(x_{1\alpha}, x_{2\beta})$ are given by¹⁶

$$\frac{i \partial \psi_{\alpha\beta}(t)}{\partial t} = [T_\alpha + T_\beta + U_{\alpha\beta} - iW_{\alpha\beta} + E(t)(x_{1\alpha} + x_{2\beta})\sin(\omega t)] \psi_{\alpha\beta}(t), \quad (4)$$

where the kinetic-energy operators T_α and T_β are represented by three-point difference formulas and the atomic potential is given by (with $Z=2$)

$$U_{\alpha\beta} = -\frac{Z}{(c_p + x_{1\alpha}^2)^{1/2}} - \frac{Z}{(c_p + x_{2\beta}^2)^{1/2}} + \frac{1}{(c_p + x_{1\alpha}^2 + x_{2\beta}^2 + 2c_r|x_{1\alpha}||x_{2\beta}|)^{1/2}}. \quad (5)$$

For the ground state of a two-electron He atom, the single-determinant mean-field approximation to Eq. (3) results in a 1D time-dependent Hartree-Fock equation given by

$$\frac{i \partial \phi_\alpha(t)}{\partial t} = [T_\alpha + V_\alpha - iW_\alpha + E(t)x_\alpha \sin(\omega t)] \phi_\alpha(t), \quad (6)$$

where the Hartree-Fock potential may be written as

$$V_\alpha = -\frac{Z}{(c_p + x_\alpha^2)^{1/2}} + \sum_a \frac{\Delta x'_a |\phi'_a(t)|^2}{(c_p + x_\alpha^2 + x_a'^2 + 2c_r|x_\alpha||x_a'|)^{1/2}}, \quad (7)$$

and Δx_a is the grid spacing.

Both time-dependent equations (4) and (6) may be propagated forward in either real or imaginary time using an implicit approximation to the time-evolution operator.¹⁶ Choosing $c_p = \frac{1}{2}$ to closely match the total energy of the real helium atom, $c_r = -1$ to maximize the correlation energy, and setting $W = E(t) = 0$, both time-dependent equations (4) and (6) when propagated in imaginary time relax initial Gaussian functions to the

ground state of the two-electron He atom. Contour maps of $\log_{10} |\psi(x_1, x_2)|^2$ in the particle coordinate plane are found in Fig. 1 for both the 2D lattice and 1D HF ground-state solutions. The Hartree-Fock energy is found to be -77.1 eV, while the 2D lattice energy is only slightly lower at -77.3 eV. The Hartree-Fock energy for the real helium atom is -77.9 eV, which represents 98.6% of the total energy.²⁵ Although correlation effects have a relatively small effect on the total energy, they do produce a noticeable pinch along the x_1, x_2 diagonals in the 2D lattice solution. The amount of spatial symmetry breaking may be controlled to some degree by varying the correlation parameter c_r between -1 and $+1$.

Once the ground state of the two-electron He atom was obtained, both time-dependent equations (4) and (6) were propagated forward in real time. Our choice of a field intensity of 1×10^{15} W/cm² and a photon energy of 9.0 eV was stimulated by a previous TDHF calculation for the three-photon ionization of helium.¹⁸ In Fig. 2 the time evolution of the total wave-function norm, $\langle \psi(x_1, x_2, t) | \psi(x_1, x_2, t) \rangle$, and the ground-state probability, $|\langle \psi(x_1, x_2, 0) | \psi(x_1, x_2, t) \rangle|^2$, are plotted for calculations using the 2D lattice, 1D frozen-core TDHF, and full nonlinear TDHF formulations. In all cases the total wave-function norm and ground-state probability decrease with time due to absorption on the boundary by the W potential. Field oscillations are seen in the ground-state probability. The 1D frozen-core TDHF solution is found by the replacement $\phi'_\alpha(t) \rightarrow \phi'_\alpha(0)$ in Eq. (7) for the Hartree-Fock potential. Since there are only two electrons present in the single shell of helium, the frozen-core TDHF solution may be more accurately described as a "single active electron" model.¹⁸ The close agreement between the 2D lattice and frozen-core TDHF methods is confirmation that the main ionization mechanism is one electron moving away shielded from the bare nucleus by the remaining electron. The spatial

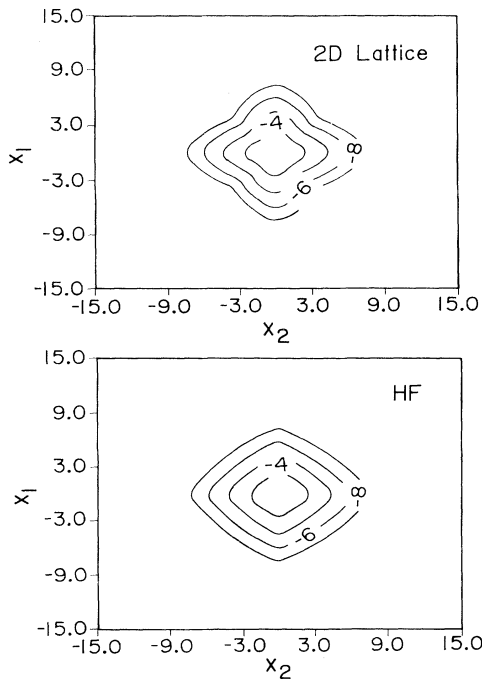


FIG. 1. Logarithm of the probability density for the ground state of "He" in a contour map. Upper plot is from a 2D lattice calculation, while the lower plot is from a 1D Hartree-Fock (HF) calculation.

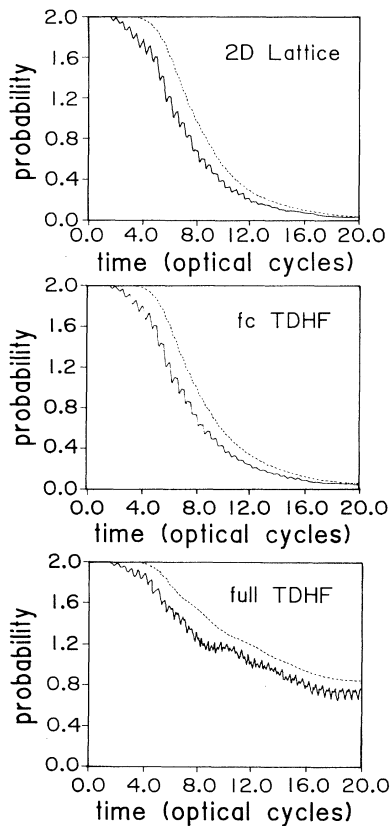


FIG. 2. Time evolution of the wave-function norm (dashed curve) and ground-state probability (solid curve) for "He." The upper plot is from a 2D lattice calculation, the middle plot is from a 1D frozen-core time-dependent Hartree-Fock calculation, while the lower plot is from a 1D full nonlinear time-dependent Hartree-Fock calculation.

asymmetry induced by correlation thus has a dramatic effect on the ionization process. The 1D full nonlinear TDHF solution preserves the spatial symmetry since the potential is continually time updated by the wave function. This represents an ionization mechanism in which both electrons move away simultaneously in the field of an increasingly bare nucleus. From the exponential decay of the total wave-function norm, an ionization rate of 6×10^{14} Hz can be extracted for both the 2D lattice and frozen-core TDHF methods. For the full nonlinear TDHF method, however, the rate of ionization decreases with time. Finally, we note that the comparison between the frozen-core and full nonlinear TDHF results for He are in qualitative agreement with the same comparison made between previous TDHF calculations for the real helium atom.¹⁸ Because of phase-space restrictions, however, the frozen-core TDHF ionization rate in the model He atom is an order of magnitude larger than its counterpart in the TDHF calculation using the full 3D electrostatic interaction.¹⁸

In conclusion, by comparing an exact time-dependent solution of a 2D model of the helium atom with the approximate TDHF method, we find that caution must be exercised when using the full nonlinear TDHF method to describe multiphoton ionization. This has important implications for future applications of the TDHF method to study collective effects in the multiphoton ionization of rare-gas atoms. We further suggest that the procedure of reducing 3D interactions to 1D interactions, coupled with the growth in advanced computing capability, makes possible the direct study of the dynamics of many types of few-body problems.

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