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Quasiclassical Approach to Ionization of Atoms by Strong Laser Pulses : the Comparison with the "Simpleman's" Models and Newtonian Dynamics.

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Abstract

We study the energy spectrum of the ionized electrons and compare it to the results of "naive" theories of multiphoton ionization of atoms (S. Augst et al., Phys. Rev. Lett., **63**, 2212, H. B. van Linden van den Heuvell, H. G. Muller, ref. 11, P. Corkum et al., Phys. Rev. Lett., **62**, 1259, (1989) D. A. Tate et al., Phys. Rev. A, **42**, 5703 (1990)). These, "naive" or "simpleman's" theories have demonstrated quite a spectacular success in the recent past, being a basis for explanation of the most observed experimental results. Our study also present a testing ground for what classical and semiclassical theories can and cannot explain.

Introduction

The current paper is a direct continuation of our previous paper [1], in which the authors proposed to study electron-electron correlations in the quasiclassical model of Kirschbaum and Wilets (also called Fermi Molecular Dynamics, the FMD) [2] with respect to multiphoton ionization. This model has been already implemented in the paper of Wasson and Koonin [3] for the study of multiphoton ionization rates for multielectron atoms. In the present paper we provide the comparison between quasiclassical FMD and purely classical models for multiphoton ionization. Here, we concentrate on the second simplest quantum system, the *He* atom.

The closest benchmark available for comparison is the set of so called "simpleman's theories", or SM's (a name suggested by Gallagher [4]). These theories become applicable for extremely high laser irradiances, where over the barrier ionization (OBI) becomes dominant [5]. This regime is characterized by the property that, at least for a part of the period of the external

field, the field becomes so strong that no bound states can exist in the combined potential of the atom and laser. As was demonstrated in Ref. [6], this regime can be connected with tunneling formulas, which are applicable for fields much weaker than atomic. A notable feature of the SM theories is that their predictions depend only on parameters of the laser pulse (intensity, frequency, and duration) and on the phase of the electron motion relative to the phase of the laser field. Another benchmark will be comparative simulation of the energy gain in the classical model of the *He* atom (Section 4).

The successful implementation of the classical and semiclassical methods in other, closely related fields of physics, such as ionization of Rydberg atoms [7], scattering problems [8], and interaction of ions with surfaces [9] prompts the use of quasiclassical computer calculations for this problem. In the pioneering work by Gaida, Grochmalicki, Lewenstein and Rzazewski, the methods of classical mechanics were used for three-dimensional simulation of the hydrogen atoms in strong fields [10].

However, in this case the purely classical methods suffer a great deficiency. They cannot provide for the stability of the systems more complicated than the hydrogen atom and hydrogen-like atoms and ions in external fields. The known stable multielectron systems have a high degree of symmetry [11] which is destroyed by the external field. Unlike collision problems, the stability of atoms should be assured for the characteristic times of thousands and tens of thousands of atomic periods even for the shortest available pulses.

Section 2. The "simpleman's" theory of the multiphoton ionization

The SM model of multiphoton ionization has its origin in ideas first expressed by van den Heuvell and Muller [12] and Corkum [6]. Both Corkum and Gallagher have performed actual experiments on real atoms which seem to have provided good evidence for the correctness of the model, within its domain of applicability.

The SM describes ionization in the regime of laser irradiances $I_0 \sim Z^6(2n)^{-8} a.u.$ for electrons bound in H-like ions with principal quantum number n and ionic core charge Z . This is the regime of so called over the barrier ionization (OBI). This regime is characterized by the property that, at least for part of the period of the external field, the field becomes so strong that no bound states can exist in the combined potential of the atom and

laser. For smaller irradiances, only stochastic ionization and tunneling are possible. In the relatively weak fields these two mechanisms are complementary: the ground state is ionized by tunneling (non-existent in the proposed model) and the highly excited states are ionized by stochastic mixing with the continuum [7].

An assumption of adiabaticity is also usually made, so that the laser frequency must be low, $\omega \ll Z^2/n^3$, which means that the electron will execute many orbits during one laser period. Finally, this essentially classical approach to ionization also requires that $n \gg 1$, if a comparison to real atomic systems is contemplated.

For linear polarization, the laser electric field takes the form

$$\vec{E}(t) = \hat{x} E_0 \sin(\omega t + \beta) \quad (1)$$

having assumed, for convenience of the ensuing analysis, an abrupt turn-on at $t=0$ with phase β . The electron is imagined to emerge into the continuum at a distinct time t' . Once having become unbound, it is considered to be unaffected by the binding potential, moving only under the influence of electric field. No possibility of recapture is admitted.

Therefore, for $t \geq t'$, the velocity of the ionized electron is just

$$\vec{v}(t) = \hat{x} \frac{E_0}{\omega} [\cos(\omega t' + \beta) - \cos(\omega t + \beta)] + \vec{v}(t') \quad (2)$$

where $\vec{v}(t')$ is the velocity at the moment of release ($|\vec{v}(t')| \leq Z/n$). The cycle-averaged kinetic energy of the unbound electron is then,

$$U(t') = [v_x(t') + \frac{E_0}{\omega} \cos(\omega t' + \beta)]^2/2 + [v_y^2(t') + v_z^2(t')]/2 + U_p \quad (3)$$

where U_p is the ponderomotive potential

$$U_p = \frac{E_0^2}{4\omega^2} \quad (4)$$

For cases in which $E_0 \sim (Z/n)^3$, it is "intuitive" to expect that ionization will occur primarily at a maximum of the driving field; i. e. when $\omega t' + \beta = \pi/2, 3\pi/2, \dots$. The cycle averaged kinetic energy of the unbound electron must be then $U(t') = v^2(t')/2 + U_p$.

In the opposite extreme, when $E_0 \ll (Z/n)^3$, the ionization should occur at the instant of turn on ($t' = 0$). Consequently, in this limit, the cycle averaged kinetic energy is $U(t') = [v_x(0)^2 + (E_0/\omega)^2 \cos^2(\beta)/2] + U_p$.

assuming no correlation between $v_x(0)$ and the driving field.

All of the preceding is in accordance with the strict SM theory. However, it is also clear that OBI can occur whenever it happens that $E(t') \geq Z^3/16n^4$. This slightly different approach to determining t' implies for the kinetic energy

$$U(t') = \frac{v^2(t')}{2} + U_p + \frac{1}{2\omega^2} [E_0^2 - \frac{Z^6}{256n^8}] \quad (5)$$

The difference between these two formulations can be great, if $\omega \ll 1$, and $E_0 < (Z/n)^3$. Numerical calculations performed for classical systems seem to validate the SM, rather than the notion of simple OBI, as described here.

If, subsequent to ionization, the driving field is turned off “slowly”, then electron kinetic energies will be modified to $U = v^2/2$, in the case that $E_0/\omega \leq (Z/n)^3$, and $U = (E_0/\omega)^2 \cos^2(\beta)/2 + v^2/2$, if $(E_0/\omega) \gg (Z/n)^3$. In either case, an energy equal to U_p will be returned to the driving field. Our numerical calculations have all been performed in this mode.

The situation for circular polarization is even simpler. If the laser electric field has the form

$$\vec{E}(t) = \frac{E_0}{\sqrt{2}} [\hat{x} \sin(\omega t) + \hat{y} \cos(\omega t)] \quad (6)$$

for $t \geq 0$, then the velocity of an unbound electron must be

$$\vec{v}(t) = \frac{E_0}{\sqrt{2}} (\hat{x} [\cos(\omega t' + \phi) - \cos(\omega t + \phi)] + \hat{y} [\sin(\omega t + \phi) - \sin(\omega t' + \phi)]) + \vec{v}(t') \quad (7)$$

for $t > t'$. The cycle averaged kinetic energy is then

$$\begin{aligned} U(t') &= \frac{v^2(t')}{2} + 2U_p + \frac{E_0}{2\omega^2} [v_x(t') \cos(\omega t' + \phi) - v_y(t') \sin(\omega t' + \phi)] \\ &\approx \frac{v^2(t')}{2} + 2 \cdot U_p \end{aligned} \quad (8)$$

since no correlation is expected between $v(t')$ and $E(t')$. And as previously, if the driving field is turned off slowly before the kinetic energy is sampled, then $U(t') = v^2(t')/2 + U_p$. For circular polarization the form of the energy spectrum of the emitted electron is invariant under changes in magnitude of the driving field E_0 or its initial phase. Finally, we note that the strict SM makes no predictions about the *rate* of photoionization, except in the very strong field domain ($E_0 \gg Z^3/16n^4$) where ionization is considered to be immediate.

However, a slight modification of the SM, suggested by Corkum, enables one to make a precise prediction of the emitted-electron kinetic energy spectrum, for linear polarization, even if the turn-on is not abrupt. Recourse can be made the well-known formula for the rate of (static) electric field ionization, R_{FI} , from the ground state of atomic hydrogen,

$$R_{FI} = \frac{4}{|F(t')/F_{at}|} \exp\left(-\frac{2}{3|F(t')/F_{at}|}\right) \quad (9)$$

generalized to a time-dependent field, where $F(t')$ may be given by Eqs. (3), (7) (but, if desired, a pulse envelope function can be included). Then, the relative probability $N(U)$ of appearing in the continuum with energy $U(t')$, where $U(t')$ is given by equation (5), is determined parametrically as a function of $F(t')$ by

$$N(U) = \frac{F_{at}}{F(t')} \exp\left(-\frac{2F_{at}}{3F(t')}\right) \quad (10)$$

for $0 < t' \leq \pi/2$. An example of the application of this semiclassical procedure appears in the following figure. Note that the formula for R_{FI} can sensibly only be applied if $F_0 < 1$. The energy spectra for Corkum's version of SM and a pulse of constant intensity are reproduced on Figs. 1a-1c. They will serve as a benchmark for a comparison with results of quasiclassical and classical simulations.

Section 3. Multiphoton ionization in a field of constant intensity

For the FMD to have some validity it is necessary that its results should be compatible with the SM theory, which has been proven so succesful in explanations of photoionization of atomic gases [5, 6, 14]. The detailed comparison, however unphysical (because all real ultrashort pulses have smooth envelopes), is the best available benchmark for our quasiclassical program, because all of the one electron phenomena can be easily interpreted in terms of SM observations.

The simulation was performed as follows. We ran statistically representative (usually several hundred trajectories) dynamical evolutions of the quasiclassical H_e in an external field with randomized initial conditions. Further, we selected some special orbit, like Langmuir or Wannier, as an initial state (both are stable in FMD approach). Then we randomize the

phase of atomic oscillations (one angular parameter), and the direction of the field with respect to the atomic orbit (two angular parameters). Naturally, the choice of quasi-ergodic initial state [10, 15, 16] will be based on more solid physical grounds. However, our simulations provide the evidence, that at least for moderate fields, the final bound state is close to quasi-ergodic.

We analyzed the data in two ways: 1) by histograms of the electronic energy, similar to the of real spectra of electronic energies in experiment, 2) by construction of “covariance maps” [17]. The values for energies, angles and angular momenta were established in aftermath of the pulse when all ionized electrons are in a free motion, while all electrons with negative energies are corresponding to some bound zero-field states. The immediate rise of the pulse doesn’t present any difficulties. To avoid computer difficulties in aftermath of the pulse, we add the gaussian tail to the back of rectangular pulse: $\exp\left(-\frac{(t-t_{imp})^2}{\Delta t^2}\right)$ with $\Delta t \sim 100a.u.$ of time. This provides smooth fall of the intensity after the end of the essential part of the pulse $t < t_{imp}$.

The energy spectrum for linear polarization and the field of the form $F(t) = F_0 \sin(\omega t)$ is given in the Fig. 2a. For the rectangular pulse the typical energy scale is provided by twice the value of the quiver energy (1250 a.u. for our case); $E_i = 2U_p$ (the results of Section 2 for the electric field stronger than atomic; see also Fig. 1), yet, there is substantial dispersion of energies below this value (see [1] and Section 4). This suggests that ionization happens almost immediately after the pulse is turned on. Then, the motion of the electron is quasi-free.

The case of circular polarization is even simpler. The energy spectrum is almost monokinetic with $E_i = 2U_p$ (Fig. 2b). The difference between linear and circular polarization can also be demonstrated through the relationship between energy and angular momentum. While for the linear polarization, the angular momentum is more or less randomly distributed in the vicinity of relatively small values, the angular momenta for the circular polarization are essentially proportional to the energy (or intensity, for that matter) with rather big proportionality coefficient of order $L_i = \frac{\hbar}{2\omega} E_i \sim 25E_i$ (Fig. 3).

The previous results for linear polarization were produced for the field proportional to $\sin(\omega t)$. “Simpleman’s” theories predict drastic dependence of the energy spectrum on the initial phase of the field. Because the electrons, under extreme diabatic conditions of excitation, are freed almost immediately, the initial phase of the electric field becomes exceedingly important. We display the results for the different initial phases for the laser field:

$\sin(\omega t + \beta)$, $\beta = 0, \frac{\pi}{4}, \frac{\pi}{2}$; other choices of $\beta = \frac{n\pi}{4}$ are similar (Fig.2,4). We see, that under strong-field conditions, for the sinusoidal field the energies are close to twice the value of the quiver energy; for the phase shift of 45° the electron absorbs $E_i \sim U_p$, and the energy spectrum for the cosinusoidal field is localized around zero.

The results of our computations, in general, corroborate the conclusions of “simpleman’s” theories of multiphoton ionization. This is a bit surprising, because these theories are based on assumptions that are, rigorously speaking, valid only for the fields weaker than the atomic field. Namely, the tunneling formulas are deduced for the tunneling exponent much bigger than unity and, consequently, small probabilities of ionization. The possibility to extend these theories into the region of very large intensities conceals some important features of correspondence between classical and quantum mechanics, which are not yet fully understood.

Section 4. The comparison between Wilets-Koonin model and purely newtonian solution for the Helium atom.

In previous section the Wilets-Koonin model for the multiphoton ionization was justified by its comparison with widely used “simpleman’s” model for multiphoton ionization.

To investigate advantages (and possible shortcomings) of the new approach, we compare the results of the Wilets-Koonin model with purely classical simulations. In a very strong field the atom and molecule are inherently unstable. The energy spectrum for a very strong field is mostly determined by dynamics of interaction between quasi-free fragments of the particle and this is precisely the type of behavior upon which the “simpleman’s” theory is based. At least under the extremely high fields the results of the Wilets-Koonin and purely Newtonian dynamics should be similar.

Two problems, however close they may seem analytically, are completely different numerically. In the case of FMD close approaches to the nucleus with small relative angular momenta (billiard ball type of collisions) are highly improbable because of emergence of large repulsive forces in this case. The trajectory in the FMD case, though, is very irregular and one needs rather precise integration to be confident in the results. In the Newtonian case the situation is quite different. The orbits, outside the close vicinity of the nucleus are smooth and this allows for big tolerance of round-off error. Vice versa, in the case of relatively small fields, the attraction of the nucleus dominates and unphysically close approaches of the electron to

the nucleus become frequent. The situation, when the particles of the opposite charge can form bound states with indefinitely small energies is typical for classical mechanics, while it is prohibited by the quantization of the angular momenta in quantum mechanics (it is statistically very improbable in our quasiclassical approach). The ionization and ejection of the electrons from these, very tightly bound states, produces occasionally some electrons with high energies even for very low fields. In very high fields, because of the large amplitude of the quiver motion, these collisions are discriminated against for purely geometric reasons and the validity of classical simulation is again restored.

To prevent artificial stiffness of the Newtonian model under moderate-to-low intensity conditions we make a cutoff of the Coulomb potential at relatively large distances from the nucleus $r_{cutoff} = 0.141 a.u.$. This falls very well into the contemporary paradigm, prescribing to work with the Coulomb potential with a smoothed core [18]. While the Newtonian model has 100 percent ionization for moderate intensities ($\sim 2.0 \times 10^{14} W/cm^2$), the quasiclassical model shows no ionization at all. This was discussed in the Section 3, where the typical critical field strength for the ionization in the model with Wilets-Koonin potential were established and turned to be around $0.145 a.u.$. At $F_0 \sim 0.45 a.u.$ the ionization becomes much faster [1], Section 3b). It happens when the strength of external field becomes comparable with the atomic field strength and the OBI from the ground state dominates. Vice versa, the purely classical helium is inherently unstable in the external field. This means that the electrons are, essentially ionized from the beginning, and later, undergo only stochastic acceleration [7] in the combination of laser and atom fields.

The energy spectrum for Newtonian model is given by the Fig. 5, and the covariance map for the $E_1 - E_2$ correlation of Fig. 6. One observes, that the single electron spectrum for the classical model is, in fact, much wider and extends to the region of low energies, as the SM spectrum Fig. 1c. A plausible explanation of this is related to the absence of the threshold in a Newtonian model. The dispersion of the ejection times in Eq. (5) is much greater for the quasiclassical case, resulting in much wider spectrum of energies.

The comparison between classical and semiclassical simulation proves very important point about purely classical models. Namely, the classical models are accurately describing the quantum dynamics of the atom in moderate fields *only if* the Coulomb potential is regularized. In the classical simulations of [19], the static magnetic field was added to regularize the

model.

Now, we can point out differences between purely classical and quantum situations. First, in classical mechanics there is a possibility of very close approach of the electron to the nucleus, with reasonably low relative angular momentum. These collisions can result in two possible effects: very sharp change in the direction of the electron, with subsequent ionization, or capture of electron by the nucleus on a very low orbit, with the binding energy much smaller than 1. a. u.. Classical mechanics makes such orbits perfectly possible, while quantum mechanics prohibits them altogether. In the second case, the ionization out of tightly bound state when field rises enough produces electrons with very high energy. Another difference is applicable for multielectron systems, while it doesn't exist for hydrogen. The multielectron atomic systems are generally unstable classically and this means that there is no threshold conditions influencing the time of ejection. This results in much broader energy features (Figs. 5-6).

Conclusion.

In the present paper we investigated the correspondence between Fermi Molecular Dynamics (FM) model for multiphoton ionization and compared it with the SM theory and purely classical simulations for the *He* atom. We observe that for the very high fields ($F \geq 5.0 \text{ a.u.}$), these theories are qualitatively similar. This is very important, because our simulation deals with two-electron atoms and field intensities in which at least some of the underlying presuppositions of the SM theories are not applicable. The robust character of OBI mechanism provides that one electron phenomena will be satisfactorily described by any of these theories, even for multielectron atoms. The phenomena of electron correlation, however manifest a significant difference related to the absence of the threshold in multiphoton ionization of a classical multielectron atom.

References

- [1] P. B. Lerner, K. LaGattuta, J. S. Cohen, Laser Physics, (to be published).
- [2] C. L. Kirshbaum, L. Willets, Phys. Rev. A, **21**, 834 (1980).

- [3] D. W. Wasson, S. E. Koonin, Phys. Rev. A, **39**, 5676 (1989).
- [4] T. Gallagher, in *The Electron: New Theory and Experiment*, eds. D. Hestenes and A. Weigartshofer, vol. 45 (Dordrecht, Kluwer Academic, Boston 1991), see also D. A. Tate et al. Phys. Rev. A **42**, 5703 (1990).
- [5] S. Augst et al., Phys. Rev. Lett. **63**, 2212 (1989).
- [6] P. Corkum, N. Burnett, F. Brunel, Phys. Rev. Lett. **62**, 1259 (1989).
- [7] G. Casati, I. Guarneri, D. L. Shepelyansky, IEEE Journ. of Quant. Electr. **QE-24**, 1420 (1988); R. Jensen, S. M. Susskind, M. M. Sanders, Phys. Rep. **201**, 1 (1991).
- [8] J. S. Cohen, to be published.
- [9] H. Ryufuku, K. Sasaki, T. Watanabe, Phys. Rev. A **21**, 7451 (1980) ; J. Burgdoerfer, P. Lerner, F. Meyer Phys. Rev. A , **44** 5674 (1991).
- [10] M. Gaida, J. Grochmalicki, M. Lewenstein, K. Rzazewski, Phys. Rev. A **46**, 1638 (1992).
- [11] J. Muller Phys. Rev. A, **45** 1471 (1992); R. Richter, D. Wintgen, J. Phys. B **23**, L197 (1990).
- [12] H. B. van Linden van den Heuvell, H. G. Muller, in *Multiphoton Processes*, (eds. S. J. Smith, P. L. Knight, Cambridge Univ. Press, Cambridge, 1989), p.25
- [13] Q. Su, J. H. Eberly, Phys. Rev. A, **43**, 2474 (1991).
- [14] G. Gibson, T. S. Luk, C. K. Rhodes et al., Phys. Rev. A, **41**, 5049 (1990); J. A. Cobble, G. T. Schappert et al., Journ. Appl. Phys. **69** 3369 (1991); L. J. Frasinski et al., Phys. Rev. Lett. **58** 2424 (1987).
- [15] J. G. Leopold, I. C. Percival, J. Phys. B, **12**, 709 (1979).
- [16] J. S. Cohen, J. Phys. B **18**, 1759 (1985).
- [17] L. J. Frasinski, K. Codling, P. A. Hatherly, Science, **246**, 1029 (1986).
- [18] Q. Su, J. H. Eberly, Phys. Rev. A, **43**, 2474 (1991).
- [19] B. Ritchie et al., Phys. Rev. A, **41**, 6114 (1990); C. M. Bowden et al., Phys. Rev. A, **46**, 592 (1992).