Strong Field Ionization in Arbitrary Laser Polarizations

M. Protopapas\textsuperscript{1}, D.G. Lappas\textsuperscript{2} and P.L. Knight\textsuperscript{1}

\textsuperscript{1}Optics Section, Blackett Laboratory, Imperial College London, London SW7 2BZ, U.K.
\textsuperscript{2}Department of Physics, Lund Institute of Technology, PO Box 118, S-221 00 Lund, Sweden

Abstract

We present a numerical method for investigating the non-perturbative quantum mechanical interaction of light with atoms in two dimensions, without a basis expansion. This enables us to investigate intense laser-atom interactions with light of arbitrary polarization without approximation. Results are presented for the dependence of ionization and high harmonic generation on ellipticity seen in recent experiments. Strong evidence of stabilization in circular polarization is found.

32.80.Fb, 32.80.Wr, 42.50.Hz
In the last ten years the interaction of intense laser light with atoms has been of great interest because of the many novel non-perturbative phenomena that have been observed, such as Above Threshold Ionization (ATI), High Harmonic Generation (HHG) and Stabilization [1]. The strongly non-perturbative nature of all these phenomena makes theoretical analysis difficult. However, considerable success has been made in using semi-analytic approaches and numerical methods for integrating the Schrödinger equation. The numerical approach has been used extensively in the past ten years due in part to the increase in available computing power, the lack of approximations (except for those inherent to the Schrödinger equation and the dipole approximation) and the rather limited applicability of analytic approaches. Despite the success of the numerical methods most investigations have dealt with only linearly polarized light: the reason for this is that at least two dimensions are needed in the case of arbitrary polarization, so established one-dimensional approaches [2] are inappropriate. Also, current three-dimensional approaches [3–6] are unable to deal with the enormous basis set needed to represent adequately the evolving wave function unless a parallel supercomputer [7] is used. The approach used by many authors [3] to describe atomic dynamics in strong laser fields expands the atomic wave function into sums of products of radial wave functions (channels) and angular factors. These channels are coupled together by the very strong laser field, and for linear polarization, the channel couplings lead to tri-diagonal sparse matrices amenable to efficient numerical solution. This approach entirely fails for polarizations other than linear, as the matrix sparseness properties totally disappear [8].

Recent experiments [9,10] on the very strong dependence of high harmonic generation on polarization ellipticity demand theoretical approaches to intense field ionization going beyond the existing methods which rely on linear polarizations. In this letter, we introduce a method for integrating numerically the two dimensional Schrödinger equation without using a restrictive basis expansion. This enables us to study light of arbitrary polarization and realistic frequencies, interacting with an atom. Fully quantum mechanical results for ionization and high harmonic generation are presented for circular and elliptically polarized
light, showing the strong ellipticity dependence seen in recent experiments \[9,10\]. Also, we show that in the intermediate regime between tunneling and multiphoton ionization the ionization dependence on ellipticity is opposite to that seen with purely tunneling dynamics; this may be critical for experiments towards generating attosecond pulses.

As in all studies of this kind the starting point is the Schrödinger equation (atomic units are used throughout):

\[
i \frac{\partial}{\partial t} \Psi(x, y, t) = \left[ -\frac{1}{2} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) - \frac{1}{\sqrt{a^2 + x^2 + y^2}} + \left( x E \sin \omega t + \epsilon y E \cos \omega t \right) f(t) \right] \Psi(x, y, t),
\]

where \(\Psi(x, y, t)\) is the two dimensional wavefunction, \(f(t)\) the pulse envelope and \(\epsilon\) the ellipticity. The second term on the right hand side is the smoothed Coulomb potential \[2\] used to avoid numerical problems associated with the singularity at \(x = y = 0\) \((a = 0.8\) so that the ground state has the same binding energy as hydrogen, i.e. \(-0.5\) a.u.). The pulse envelope, \(f(t)\), used here is a 2-cycle linear turn-on followed by 2 cycles of constant intensity and then a 2-cycle linear turn-off, which ensures no residual drift motion in the free electron displacement. The ellipticity is defined such that \(\epsilon = 0\) represents linear polarization along the x-axis and \(\epsilon = 1\) circular.

Equation 1 is integrated using a method analogous to that implemented by Grobe and Eberly \[11\] and others in their studies of one-dimensional two-electron systems in which both electrons are restricted to one dimension. In our case there is only one electron with two degrees of freedom. The core of the approach is the split step method \[12\] in which the time evolution operator is split into kinetic and potential operators:

\[
U(\Delta t) = \exp[-iT\Delta t/2] \exp[-iV\Delta t] \exp[-iT\Delta t/2] + O(\Delta t^3),
\]

where \(T\) is the kinetic part of the Hamiltonian (including the dipole interaction, in a ‘minimal coupling’ form) and \(V\) the potential part. The action of the kinetic operator is efficiently carried out in Fourier space, while the action of the potential operator is carried out in real space. An absorber is used to remove any part of the wavepacket reaching the boundaries,
so that artificial reflections are avoided. It is assumed that any wave function reaching the end of the grid box represents ionization, and so the remaining norm gives the necessary information about the amount of ionization occurring. This means that any population in high lying Rydberg states may also be removed; however, this is negligible for the parameter regime of interest here, as we have verified by varying the box dimensions. In all the cases presented, the initial wave function is the ground state, found by imaginary time integration. A more detailed discussion on various computational aspects can be found in ref. [11].

In Fig. 1 we show the wavepackets generated after 3.5 cycles of evolution for the previously described pulse shape with laser light of wavelength 526 nm ($\omega = 0.0867$ a.u.), i.e., the wavelength of a frequency doubled Nd:YAG laser and intensity $3.51 \times 10^{14}$ W cm$^{-2}$ (0.01 a.u.). For the case of linear polarization it is clear that the wavepacket is concentrated about the polarization axis, but there is a non-negligible width due to transverse wavepacket spreading. Interestingly, side lobes can be seen in the wavepacket; these are formed by a combination of the slow transverse spreading, the relatively fast laser driven motion in the polarization direction and the Coulomb attraction (rescattering).

The behaviour of the wavepacket created by circular polarization is very different. The packet resembles a spiral with modulations within the tail, formed as the wavepacket streams away from the atom down the potential barrier in the radial direction. As well as being interesting in their own right the wavepackets show pictorially that the recollisions necessary for efficient HHG can only occur for light with $\epsilon \approx 0$ [9]. The complex structure of the wave function in the linear polarization case reflects the fact that rescattering from the atomic core, according to the recollision model of HHG [13–15], produces strong time-dependent interference patterns in the vicinity of the atomic core, which are responsible for much of the structure in the spectrum of HHG [16]. Those interferences are absent in the circular polarization case, as is clear from Fig. 1(b), due to the fact that the probability of rescattering with the atomic core is negligible. We have observed a smooth transition from the recollision behaviour resulting from linear polarization to the swirling tail of the circular polarization, by integrating the wavepacket dynamics for ellipticities ranging from 0 to 1.
This fact can be shown explicitly by investigating the dependence of ionization and HHG spectra on ellipticity. This is shown in Fig. 2: in (a), for a fixed intensity of 0.01\,a.u. the normalization at the end of the pulse is plotted as a function of ellipticity, where the results have been scaled such that the normalization for the linear case ($\epsilon = 0$) equals unity. One has to keep in mind that since the intensity is kept constant for varying ellipticity, the amplitude of the electric field in the initial (linear) polarization direction changes. The reduced ionization in the initial direction is compensated by the possibility of electron ejection in all other directions. The result is a net increase of total ionization yield, especially for such high intensities that Over the Barrier Ionization (OBI) occurs. This effect is even greater if equal electric field amplitudes are compared rather than equal intensities. In this case the probability of ionization is obviously enhanced, as the electron can escape from the atom by tunneling through or over the barrier at any time in the direction of the instantaneous electric field.

Clearly, for these parameters we find that as the ellipticity is increased the amount of ionization increases by a factor of approximately 3. Past experiments [17,18] have found that the threshold for ionization is higher for circular polarization. However, these experiments have been carried out in the tunneling regime where standard tunneling theory [19] predicts higher ionization rates for linear polarization. The results presented here are not strictly within the tunneling regime (due mainly to the relatively high frequency) and represent new behaviour. In Fig. 2 (b), we have used the same parameters as in Fig. 1 (the spectra are rescaled such that the fundamental peak is equal to one in intensity for each ellipticity), and now find that the harmonics act as seen in experiments [9,10]: as the ellipticity is increased, the harmonics become less intense until no harmonics are discernible from noise. Only the first 4 harmonics can be seen for $\epsilon = 0.6$, and the peaks are found to be rather broad, indicating that they are generated on a short timescale. In addition there seems to be a correlation between the ionization behaviour and the ellipticity beyond which the harmonics are destroyed: the normalization flattens out after approximately $\epsilon = 0.7$ which is also the ellipticity at which the harmonics disappear.
To highlight the very different behaviour of the ionization for various ellipticities we have calculated the normalization at the end of the pulse for linear and circular polarization as a function of intensity (Fig. 3). At low intensities we see the expected perturbative multiphoton behaviour, observed in experiment [10], that linear polarization produces a greater amount of ionization because of the transition rule restrictions on circular polarization transitions. However, as the intensity is increased to the critical intensity for over the barrier ionization (around 0.005\textit{a.u.} of intensity) we observe that the behaviour changes such that circular polarization becomes more efficient in producing ionization and indeed for much of the intensity range shown it is at least an order of magnitude greater. Note that the normalization curve for linear polarization is not smooth: this was previously observed by Pindzola and Dörr [20] and LaGattuta [21] and assigned to AC Stark shifted resonances and threshold effects. However, we find, as in ref. [21] that for circular polarization these structures do not occur, perhaps because of the more restrictive circular dipole selection rules, at least in the perturbative regime of intensities.

Finally, we observe a minimum in the normalization implying that stabilization is occurring. This is in agreement with calculations made by Pont and Gavrila [22] for circular polarization using time-independent high frequency Floquet theory. The lowest frequency which they have used is $\omega = 0.125 \text{ a.u.}$, while we have found stabilization, albeit not to the same degree, for a frequency 1.5 times smaller. Interestingly, for the same parameters linear polarization shows no signs of stabilization. This may be due to the atom not surviving the passage through ‘death valley’ [23]. We are currently investigating adiabatic stabilization with much higher frequencies and intensities, the results of which will be presented elsewhere.

In summary, we have investigated numerically, with two-dimensional ab initio calculations, the dependence of ionization and HHG on ellipticity. In particular, we have found that the harmonics are destroyed by elliptical light, as seen in recent experiments [9,10]. In a parameter regime where the tunneling picture is not entirely valid, circular polarization produces a greater amount of ionization than linear. We have also found evidence
for stabilization in circularly polarized light in qualitative agreement with previous time-independent calculations. The method we propose opens up the possibility of studying fully quantum-mechanically wavepacket evolution in time-dependent ellipticities proposed for the generation of ultrashort pulses of harmonics [24]. As well as this exciting prospect, the unified description of the strong-field phenomena, based on the exact wavepacket dynamics presented here, will open the way to resolve problems associated with, e.g., the accuracy of the ‘two-step’ model for harmonic generation, time-dependent laser polarization, control of harmonic generation in time, ionization with elliptical light, elliptical stabilization of wavepackets, angular distribution of photoelectrons in ATI, and many other important questions in strong-field physics that could be addressed only by approximate and speculative theoretical models in the past.

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REFERENCES


[8] Huens et al [7] have been able to circumvent, to some degree these problems by using Coulomb-Sturmian basis functions, but are able to study cases with high frequency only.


FIGURES

FIG. 1. Snapshots of the probability density taken after 3.5 cycles for (a) linear polarization and (b) circular polarization with intensity $3.51 \times 10^{14} \text{Wcm}^{-2}$ and wavelength 526 nm. Distances (X,Y) are shown in atomic units (a.u.).

FIG. 2. (a) Rescaled normalization at the end of the pulse as a function of ellipticity for a 6 cycle pulse with maximum intensity $3.51 \times 10^{14} \text{Wcm}^{-2}$ and wavelength 526 nm. (b) Rescaled HHG spectra as a function of ellipticity for the same laser parameters.

FIG. 3. Normalization as a function of peak laser intensity for a 6 cycle pulse with wavelength 526 nm, for linear and circular polarization.