Identification of states responsible for ATI enhancements in argon by their calculated wave functions

H.G. Muller

FOM-Institute for Atomic and Molecular Physics,
Kruislaan 407, 1098 SJ Amsterdam, The Netherlands.

Abstract: Electronic wave function movies obtained by numerically solving the time-dependent Schrödinger equation are used to elucidate the mechanism responsible for enhancements in the ATI spectrum of argon between 30 and 40 TW/cm^2.

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Photo-ionization of argon by 800-nm light is a complicated quantum-mechanical process [1] that can only be described by non-perturbative methods. In particular, electrons of many different energies are emitted, since ionization can be accomplished by absorption of many different numbers of photons. This phenomenon is known as above-threshold ionization, (ATI[2]), and leads to a sequence of peaks in the electron spectrum, spaced by the photon energy $\hbar\omega$. As usual for the photo-electric effect, the kinetic energy of the released electrons is equal to the energy in the absorbed photons minus the ionization potential. The latter, however, varies by a large amount as a function of intensity, so electrons of any energy can appear[3]. But electrons do not appear at any energy in equal amounts: the electron emission is resonantly enhanced in a way that makes it very sharply dependent on intensity.

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Recently, it was shown that numerical solution of the time-dependent Schrödinger equation for this process in the single-active-electron approximation[4, 5, 6] is capable to generate electron spectra[7, 8] that show superb agreement with experiments[9]. The spectra themselves, however, show many unassigned or unexplained features. The calculations can be illuminating in this respect, since they allow analysis at a level of detail that is is not yet possible experimentally. In particular, it is possible to study charge densities as a function of time with sub-atomic resolution.

In this paper, we present wave functions obtained by such calculations. The Schrödinger equation

$$\partial t\Psi = (\frac{1}{2}p^2 + V + \mathbf{A}(t)\cdot\mathbf{p})\Psi$$

for a single electron in three dimensions is numerically integrated with a potential modelling the argon atom[7]

$$V(r) = -(1 + 5.4\,e^{-r} + 11.6\,e^{-3.682r})/r.$$

Due to cylindrical symmetry with respect to the direction of the vector potential $\mathbf{A}(t)$ (which is fixed in space for linear polarization) the problem reduces to a two-dimensional one. This is solved in a spherical volume of radius 120 Bohr, with an absorbing mask gradualy eating the wavefunction in a reflectionless way when it gets farther than 60 Bohr from the nucleus.

At the intensities required for ionization, all states other than the ground state (which for argon has the active electron in a $3p$ state) are driven into a strong quiver motion by the laser[10], and the wave functions are thus highly dynamic. This quiver motion contributes its cycle-average kinetic energy (known as the ponderomotive energy U_P) to such states, which as a result can shift their energy by several multiples of $\hbar\omega$. The wave functions presented here are quasi-stationary states: they are periodic with the laser period, except for an overall phase factor (not affecting any observables) and some exponential decay. In an electromagnetic pulse containing a segment of constant amplitude E_o, (a 'flat-top pulse') such a state develops after transients due to the turn-on have decayed.

The wave functions are heavily dominated by the ground-state at all intensities studied: typical ionization rates are only around 0.1% per optical cycle. Only in the region of the ground state are the gradients of the Coulomb potential large enough to resist the pull of the laser. Whatever population leaves this region is driven by the laser into a strong quiver motion. It can be distinguished in a free and a bound part on the basis of its cycle-average ('drift') velocity. The bound part has drift velocities so low that it cannot escape the long-range pull of the Coulomb potential, and as a consequence it stays in the vicinity of the atom for several cycles. Due to interference of contributions from a number of cycles, it can resonantly build up to a large amplitude, strongly affecting the ionization process.

Since the large ground-state population is very compact, its charge density exceeds that of the other contributions to the wave function by many orders of magnitude, and as a result it overflows all of the presented plots. The dominance of the ground state in the plots is not much of a hindrance in identifying the resonant part of the wave function, since it is confined to a small region. Electron density emitted by the ground state into the continuum, however, first travels through the region in which charge density is building up resonantly. The momenta of the two populations are different, and this results in rapidly changing interference patterns, which might make it hard to observe the true flow of population.

The electron spectrum as a function of intensity shows many resonance enhancements (Fig. 1). Not all ATI peaks in the spectrum are enhanced by the same amount. Some

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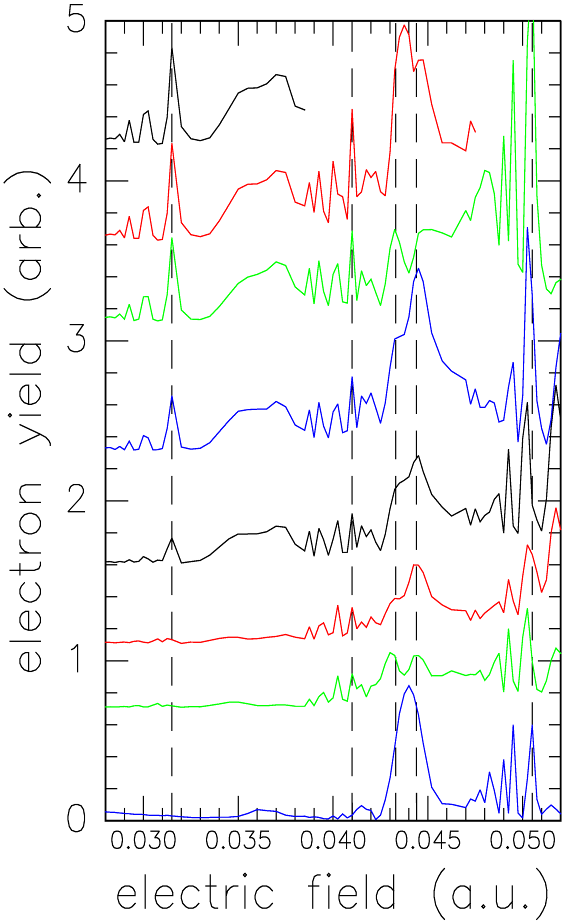

Fig. 1. Calculated partial yield of the individual ATI channels (in the polarization direction) as a function of the field amplitude E_0, divided by e^{220E_0} to supress the huge increase with intensity, and thus make representation on a linear scale possible. The curves are offset from each other for clarity. The upper seven curves represent (top to bottom) ionization by 12 to 18 photons. The lowest curve represents 28-photon ionization, and is multiplied by 10 compared to the others. The intensity range shows resonances with the g Rydberg series (left), the f and h series (middle) and again the g series (right) with 11, 12 and 13 photons, respectively. The dashed lines indicate intensities where movies are available.

enhancements are effective only in the low-order part of the spectrum, others only in a limited range of high ATI orders[11]. The remainder of this paper will present and analyze the time-dependent wave functions for some of the most prominent enhancements.

In all cases the plotted quantity is $\sqrt{10+\rho^2}\,|\Psi|^2$, where ρ is the distance to the axis of cylinder symmetry. This way of plotting gives a proper representation of the total amount of charge for large ρ, (where the square root approaches the volume element $\rho d\rho$ of a cylindrical coordinate system), thus preventing the 'disappearence' of wave function features moving away from the axis due to their spreading in the unrepresented dimension. At the same time it is able to show what happens on the axis itself (where the true volume element would vanish).

The movie linked to Fig. 2 shows the quasi-stationary state as it develops at $E_0 = 0.0315$, for linearly polarized light. This is a state that causes enhancement only in low-order ATI peaks. The charge density in a plane through the polarization axis consists of a ring of 8 roughly equally sized lobes. Only the lobes on the polarization axis are really that; due to the cylindrical symmetry, the off-axis 'lobes' in the movie are really rings

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around the polarization axis. The charge distribution is driven into a quiver motion by the laser (with an amplitude $\alpha_0 = 9.5$ Bohr), but otherwise looks identical to that of the $5g$ (m=0) Rydberg state of the unperturbed atom. The binding energy of this state is indeed such that at the quoted intensity it should be in 11-photon resonance with the ground state (assuming ponderomotive shifting).

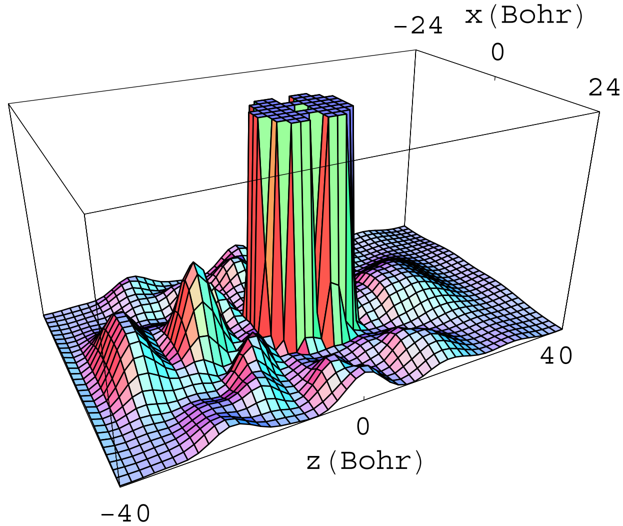

Fig. 2. One-cycle movie (1.5 MB) of the wavefunction at $E_0 = 0.0315$ a.u., where the $5g$ state is shifted in 11-photon resonance with the ground state. At this intensity the quiver amplitude is 9.5 Bohr, which brings the wave function (radius 25 Bohr) very close to the point where an electron tunneling out of the atomic ground state would appear with zero kinetic energy (16.5 Bohr).

The ground state can be seen to strongly polarize (actually we are looking at its very low-level tails of it on this scale), which eventually results in the emission of a doubly peaked wave packet. Since the displacement of a free or nearly free electron is 180^o out of phase with the force on it, this wave packet appears at the down-field side from the nucleus just as the on-axis lobe of the quivering g-state approaches the nucleus from that side, and nicely coalesces with it. The resonant intensity is special in that this merging goes accompanied with maximally constructive interference. The phase of the emerging wave packet with respect to the g-state is determined by the energy difference between the states, and is thus strongly intensity dependent through the relative ac Stark shift. At other, nearby intensities a similar wave packet would emerge from the ground state, but any attempt to build up population in the $5g$ state is frustrated, since later packets destroy population built up earlier, by destructive interference.

The $5g$-state itself also does not easily decay; the quiver motion is not large enough to drive it against the nucleus, and only the inward tails of it (penetrating the centrifugal barrier) hit the nucleus at low speed, resulting in some back-scattered low-energy free electrons. As a result the resonance enhancement caused by this state is very narrow, and affects only the low-energy electrons. In addition the state is never very much deformed (apart from the quiver displacement) compared to an unperturbed atomic state.

In the movie linked to Fig. 3 an even narrower resonance at $E = 0.0410$ is shown. Everything said about the $5g(11)$ resonance is valid here as well, except that there is

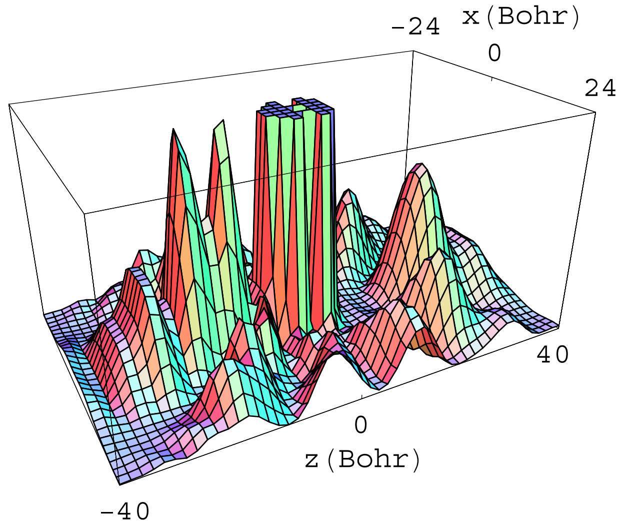

Fig. 3. One-cycle movie (1.6 MB) at $E_0 = 0.0410$ a.u., where the $n = 6$ Rydberg manifold is shifted into 12-photon resonance with the ground state. By its number of angular nodes, the wavefunction can be identified as a nearly pure $6h$ state.

one extra nodal plane, (and consequently the wave function has opposite parity), and the intensity corresponds to a ponderomotive shift that brings $n = 6$ states into 12-photon resonance, i.e. this is the $6h(12)$ resonance.

Where the $5g(11)$ resonance could stay clear of the nucleus, the $5g(13)$ resonance is not so lucky. The 13-photon resonance happens at $E = 0.0505$, and at this intensity the quiver amplitude has grown to $\alpha_0 = 15$ Bohr. This is close enough to the unperturbed

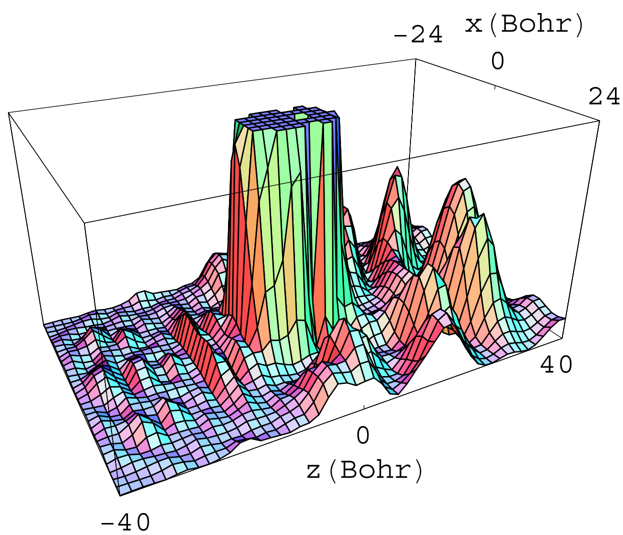

Fig. 4. Again a resonance with the $5g$ state, this time by a 13-photon transition at $E_0 = 0.0505$ a.u.. At this intensity the quiver amplitude is large enough to make the state hit the nucleus, and cause some backscattering products (the short wavelength ripples that move out fast when the remainder of the wave function is at rest). (Movie size 1.5 MB)

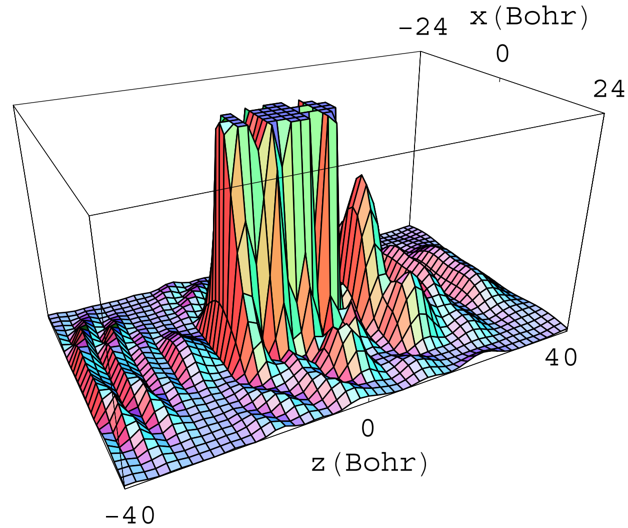

Fig. 5. One-cycle movie (1.6 MB) at $E_0 = 0.0440$ a.u., where maximum enhancement occurs in the 28th order ATI peak. A burst of high-energy electrons can be seen to move out when most of the charge is on the other side of the atom.

radius of the state for the on-axis lobe to collide with the nucleus. This results in a lot of backscattering products, that in the movie in Fig. 4 can be seen as short-wavelength ripples that speed out of the plotted region at a time when the resonant part of the wave function is turning around at the other side of the atom. The next half cycle of the light will accelerate such electrons to quite high energy[12], and indeed $5g(13)$ shows up as a very important resonance in the high-order ATI peaks. The situation is quite different for the enhancement at $E_0 = 0.0440$ in Fig. 1, shown in the movie behind Fig. 5. At this intensity there is maximum enhancement in the high-order spectrum (around order 27). In this case the atom spits out a sequence of four to five charge bunches each half-cycle. The bunches are separated by very deep minima. The first two bunches escape directly to infinity, but the next two fall back to the ion. The fourth bunch impacts on the ion at significant speed, and (partly) reflects from it. The reflected products overtake the second bunch (which is moving towards the nucleus at that time), resulting in some interference ripples at the difference of their wave vectors. In the end the second bunch is pushed outward by the field again before it hits the nucleus, as the first bunch of the next train appears and nearly grabs its tail.

In Fig. 1 this resonance seems a barely resolved doublet in the low-order ATI peaks. Indeed the resonant charge density that develops is dependent on the exact intensity used to excite it. The upper movie in Fig. 6 shows the result of excitation at $E_0 = 0.0433$. In this case two strong off-axis lobes (that is, rings) are visible, but they are much closer to the nucleus than in Fig. 2 or 3. The lobes do not seem entirely quasi-periodic; they seem to move away slightly in the radial dimension. Nevertheless, a new one that appears still overlaps part of the old one, causing enhancement by constructive interference (at this intensity). The wave function at least temporarily resembles the $4f$ state a little.

This resemblance disappears for excitation at $E_0 = 0.0444$, shown in the lower movie of Fig. 6. In this case the majority of the charge is in the on-axis lobes; two of those, have zero drift momentum and oscillate through the nucleus. The nodal plane between the lobes (which can be seen clearly only after removal of the ground state population as in [8]) identifies it as an odd parity state. Although calling it a p-state stretches the imagination, no other odd-parity states are expected at this high binding energy.

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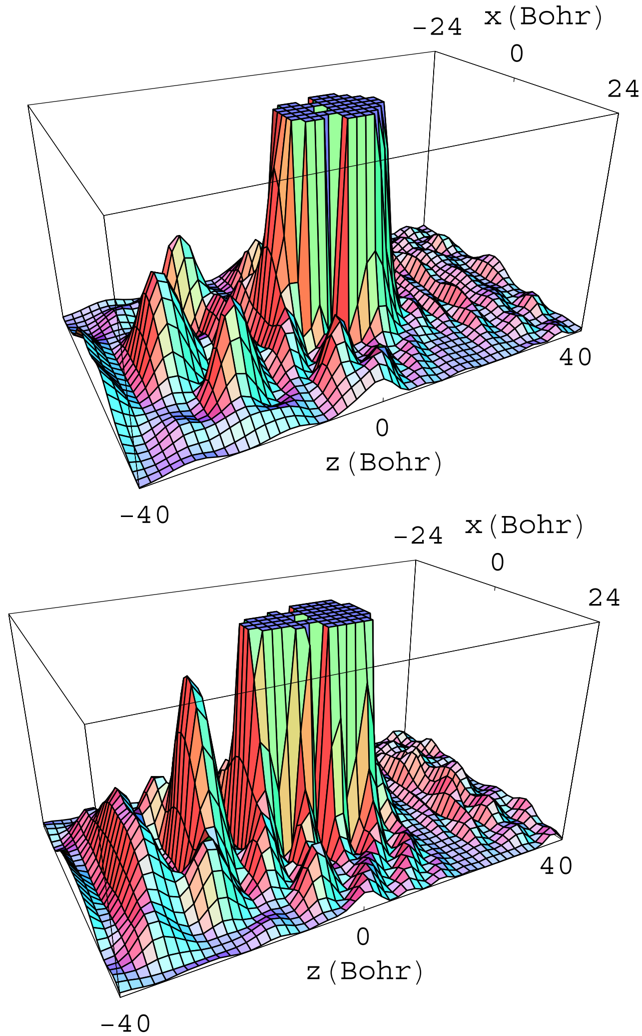

Fig. 6. One-cycle movies at $E_0 = 0.0433$ a.u. (top, 1.6 MB) and $E_0 = 0.0444$ a.u., (1.8 MB) two intensities in the opposite wings of the strong high-order enhancement. The cover frame of both movies is taken at the same phase of the laser. Note the strong off-axis lobe in the upper movie, as compared to the lower one.

Neither is the difference between the situations of Fig. 6 absolute; both show similar features, just to a different extent. This is to be expected, though, since we are dealing with overlapping resonances that can not be excited in isolation. Both states in Fig. 6 have significant lobes on the axis, and these lobes pass through the nucleus at comparatively high quiver velocity. As a result, there are energetic backscattering products, and these are responsible for the enhancement in the high-order ATI spectrum.

In conclusion, we remark that the presented electron wave functions show many small interesting details, that only become apparent after intense viewing of the respective movies.

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