High-order above-threshold ionization of argon: Plateau resonances and the Floquet quasienergy spectrum

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The Floquet quasienergy spectrum of argon in a strong laser field of 800 nm wavelength is calculated for intensities up to 7×10^{13} W cm^{-2}, and beyond for some states, using a discrete complex basis set. Many of the dressed excited states of interest shift nonponderomotively in complicated ways but keep an ionization width narrow enough to produce sharp enhancements of above-threshold ionization (ATI) through Stark-shift-induced resonances. The quasienergy map is compared to high-resolution ATI spectra for 120 fs Ti:sapphire pulses [Nandor *et al.*, Phys. Rev. A **60**, R1771 (1999)]. The plateau enhancements happen at intensities where the dressed ground state is in resonance or in the wing of resonances with dressed excited states. The resonant dressed states are identified. In many cases, the same state is responsible for an enhancement of ATI in the low as well as the high orders. No evidence is found for enhancements that are not concomitant with any curve crossing and could thereby be interpreted as channel-closing enhancement.

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I. INTRODUCTION

The above-threshold ionization (ATI) spectrum of atoms exposed to intense laser pulses normally decreases in magnitude after the first few orders, levels off in a plateau, and then drops to noise level [1]. The structure of the spectrum can be readily explained by a simple classical model, the "simple-man model": the plateau is formed by photoelectrons that rescatter from their parent ion before escaping, while those that do not rescatter (the majority) contribute only to the low ATI orders [2]. It has been known from some time that the emission of low-energy electrons is strongly enhanced, and often completely dominated, by series of multiphoton resonances between the ground state and excited states Stark shifted by the field [3]. More recently, detailed measurements of ATI in argon and xenon have revealed that the plateau part of the spectrum is also dominated by narrow resonancelike structures appearing at specific intensities, at least for pulse durations not shorter than those used in these experiments (120 fs) [4–6]. High-precision calculations on one-electron, three-dimensional models of argon and helium have revealed that these enhancements occur at particular intensities where the atom is in a state akin to a superposition of the ground state and of one or several excited states, and in which the electron, driven back and forth by the field, has a high probability of recolliding with the ionic core and being scattered in the polarization direction [6–13]. This finding indicated that the plateau enhancements originate from Stark-shift-induced resonances, like the enhancements previously found in the low-energy part of the spectrum. However, not all the resonant states were identified with particular field-free bound states.

Excited states are ignored in the simple-man model and in its semiclassical counterpart [1]. Instead of resonances with excited states, the semiclassical calculations predict the existence of plateau enhancements at particular intensities where a multiphoton ionization channel closes due to the Stark shift of the initial state with respect to the continuum threshold [14]. These enhancements can be ascribed to photoelectrons which linger for a long time in the vicinity of the core before being scattered and ejected with a large drift momentum. This mechanism differs fundamentally from the one outlined above only in that ordinary Stark-shift-induced resonances depend on an interaction between the initial state and a resonant excited state, while channel-closing induced enhancements do not. They have been found in full quantum mechanical calculations on the multiphoton detachment of electrons from a zero-range potential, which does not support any excited state [15]. They may have been observed in detachment from one-dimensional potentials, too; however, it is possible that these structures were ordinary Stark-shift-induced resonances with light-induced states lying very close to the continuum threshold [16]. Channel-closing enhancements have been proposed as an alternative explanation of the plateau resonances in rare gases: they would occur in this case not when an actual multiphoton channel closes—none have been found so far at these intensities—but when the ground state crosses an effective continuum threshold lower than the actual threshold [14].

The question of the origin of plateau resonances in rare gases has remained topical, given the current interest in the role played by the electron-core interaction in recollisional processes [17] and for multiphoton processes in ultrashort pulses. One may indeed wonder to what extent the dephasing of slow electrons due to their interaction with the core might affect the building up of channel-closing enhancements, and how these enhancements and the resonances with excited states evolve as the pulse duration is gradually reduced [14,18,19]. A better understanding of plateau resonances in long pulses would be useful on both fronts. In this work, our main aim is to shed more light on their origin by comparing existing high-resolution ATI spectra of argon to a newly

compiled Floquet quasienergy map of this atom.

For relatively long pulses such as the 120 fs Ti:sapphire pulses we are considering here, it is useful to analyze the dynamics of the atom in terms of an interplay between dressed states. These generalize the concept of stationary states of an unperturbed atom to quasistationary states of an atom exposed to a monochromatic laser field [20]. They are described by state vectors of the Floquet form $|\Psi(t)\rangle = \exp(-i\epsilon t/\hbar)|F(t)\rangle$, where $|F(t)\rangle$ is periodic in time with the same period as the field and ϵ, the quasienergy, is complex for bound states undergoing photoionization. Stark-shift-induced resonances occur at intensities where the real part of the quasienergy of the dressed ground state matches the real part of the quasienergy of a dressed excited state within an integer multiple of the photon energy [21–24]. As has been demonstrated in one-dimensional models [16], high-order ATI can be enhanced by such resonances.

We calculate the quasienergy spectrum for a wavelength of 800 nm and intensities up to 70 TW cm^{-2} (and beyond for selected states). The quasienergies are obtained as complex eigenvalues of the complex-rotated Hamiltonian represented on a discrete basis of Sturmian functions and spherical harmonics [24–27]. To the best of our knowledge, this is the first comprehensive compilation of the Floquet map of a complex atom in the visible or in the infrared [28]. We present this spectrum in some detail, given that the wavelength and the species are of experimental interest, that the precise location of each crossing is important for identifying plateau resonances, and that its main features are generic. Many of the states shift in complicated ways through a plethora of avoided crossings, changing character as the intensity increases. The assignment of resonances to field-free states is therefore fraught with difficulties.

Channel-closing enhancements should have no counterpart in the quasienergy spectrum, since they do not involve excited states. However, we find that all the plateau enhancements observed in the experimental ATI spectrum and in the time-dependent calculations are associated with quasienergy curve crossings, albeit loosely in some cases. Even at high intensity, the widths of the relevant resonant dressed states are not so large that the resonance structures are washed away.

The calculation of the quasienergy spectrum is outlined in Sec. II. The results are presented in Sec. III, and conclusions are drawn in Sec. IV. The low-energy part of the ATI spectrum at intensities between 50 and 66 TW cm^{-2} is discussed in the Appendix.

II. THEORY

As in Ref. [6], we describe the atom by the one-electron model potential

$$W(r) = [1 + Ae^{-Br} + (17 - A)e^{-Cr}]/r \tag{1}$$

in atomic units, where $A=5.25$, $B=0.97$, and $C=3.7131$ for the series converging to the $3p^5\,{}^2P^o_{3/2}$ threshold, and $A=7.3$, $B=1.15$, and $C=4.5823$ for the series converging to the $3p^5\,{}^2P^o_{1/2}$ threshold. Apart from the deeply bound $1s$, $2s$, $2p$, and $3s$ states, which have no counterpart among the valence states of the real atom, and the $4s$ state, for which there is a 2% difference, the energy levels of the model practically coincide with the centroids of the experimental fine-structure multiplets [29].

We assume that the field is linearly polarized in the z direction and write the Hamiltonian as

$$H(t) = H_0 + i\hbar\frac{e}{m}A(t)\frac{\partial}{\partial z} + \frac{e^2}{2m}A^2(t). \tag{2}$$

In this equation, H_0 denotes the field-free Hamiltonian of the model atom, $A(t)$ the vector potential of the field, and m and e the mass and the absolute charge of the electron. We set $A(t)=(F_0/\omega)\sin(\omega t)$. Within the dipole approximation, the term proportional to $A^2(t)$ has no influence on the photoionization dynamics and can be removed from $H(t)$ by a simple gauge transformation [30]. In the following, we use the transformed Hamiltonian

$$H'(t) = H_0 + i\hbar\frac{e}{m}A(t)\frac{\partial}{\partial z}. \tag{3}$$

Apart for an overall intensity-dependent shift of all levels by $-U_p$, where $U_p=e^2F_0^2/(4m\omega^2)$, $H'(t)$ has the same quasienergy spectrum as $H(t)$ [23]. (U_p is the ponderomotive energy, i.e., the cycle-averaged energy of a free electron with zero drift velocity oscillating in the field.) The energy of a continuum state lying just above the ionization threshold is equal to U_p when calculated using $H(t)$. It therefore varies with the intensity. By contrast, this energy is 0 when calculated using $H'(t)$, because of the general shift of all energy levels by $-U_p$. The position of the ionization threshold defined by $H'(t)$ thus remains the same at all intensities. The real part of the quasienergy of the high Rydberg states also remains (almost) constant as a function of the intensity, while the real part of the quasienergy of the ground state shifts downward by approximately $-U_p$. The effective binding energy of the ground state thus increases with intensity. However, the difference in energy between any two dressed states is the same whether one uses $H(t)$ or $H'(t)$. The drift kinetic energy of a photoelectron detached by absorption of N photons from a state with quasienergy ϵ is simply $\mathrm{Re}\,\epsilon + N\hbar\omega$ if ϵ is calculated using $H'(t)$.

To obtain the quasienergies, we seek solutions of the Schrödinger equation of the form [30]

$$\Psi(\mathbf{r},t) = e^{-i\epsilon t/\hbar}\sum_N e^{-iN\omega t}\mathcal{F}_N(\mathbf{r}). \tag{4}$$

The index N characterizes the net number of real or virtual photons absorbed by the atom. Making this ansatz and equating the terms oscillating as the same power of $\exp(i\omega t)$ in the Schrödinger equation yields the following system of coupled time-independent equations:

$$(\epsilon + N\hbar\omega - H_0)\mathcal{F}_N = V_+\mathcal{F}_{N-1} + V_-\mathcal{F}_{N+1}. \tag{5}$$

The operators V_+ and V_- are defined by the equation

$$i\hbar\frac{e}{m}A(t)\frac{\partial}{\partial z}=V_{+}e^{-i\omega t}+V_{-}e^{i\omega t}. \quad (6)$$

Equation (5) must be solved for the harmonic components $\mathcal{F}_N(\mathbf{r})$ and the quasienergy ϵ. For a solution to represent a bound state dressed by the field and undergoing photoionization, the harmonic components must be regular at the nucleus and must behave at asymptotic distances as superpositions of outgoing waves in the open channels and damped waves in the closed channels [31]; namely,

$$\mathcal{F}_N(\mathbf{r})\sim\sum_M f_{MN}r^{-i\gamma_M}e^{ik_M r}/r, \quad r\to\infty, \quad (7)$$

with $\gamma_M=me^2/(4\pi\epsilon_0\hbar^2k_M)$ and

$$k_M=[(2m/\hbar^2)(\epsilon+M\hbar\omega)]^{1/2}. \quad (8)$$

The branch of the square root function is chosen so that $\mathrm{Re}\,k_M>0$ for $\mathrm{Re}\,\epsilon+M\hbar\omega>0$ and $\mathrm{Im}\,k_M>0$ for $\mathrm{Re}\,\epsilon+M\hbar\omega<0$. These Siegert boundary conditions make the quasienergy complex: If E is the energy that ϵ reduces to in the zero-field limit, Δ the ac Stark shift of the state, and Γ its photoionization width, then $\epsilon=E+\Delta-i\Gamma/2$.

We calculate the quasienergies by expanding the harmonic components on a discrete basis of spherical harmonics and complex radial Sturmian functions [24–26]. The resulting eigenvalue problem for the quasienergy and the coefficients of the expansion is solved by Arnoldi iterations. Spurious solutions arising from numerical inaccuracies and solutions representing approximate continuum states rather than the relevant decaying bound states are eliminated by ignoring the quasienergies that are not stable with respect to reasonable variations of the parameters of the Sturmian functions. (The Sturmian basis is inadequate for representing dressed continuum states.)

III. RESULTS AND DISCUSSION

A. Quasienergy spectrum

All our calculations have been carried out for a field of 800 nm wavelength ($\hbar\omega=1.55$ eV). The spectra for the two core symmetries of the excited states ($J=1/2$ and 3/2) are very similar. The only important difference is that the energy levels with $J=1/2$ are about 0.18 eV higher with respect to the ground state, which has the consequence that the Stark-shift-induced resonances between the dressed ground state and most dressed excited states occur at intensities weaker by about 3 TW cm^{-2} for $J=1/2$ than for $J=3/2$. However, with rare exceptions, the energy of the photoelectrons emitted at these resonances is practically the same for both symmetries.

The $J=3/2$ spectrum between 0 and 70 TW cm^{-2} is displayed in Fig. 1. Each dot represents a calculated quasienergy. To facilitate the discussion of the results, each value of $\mathrm{Re}\,\epsilon$ is plotted shifted upward by an integral multiple of the photon energy, $N\hbar\omega$, with N chosen such that $0\le\mathrm{Re}\,\epsilon+N\hbar\omega\le\hbar\omega$. $\mathrm{Re}\,\epsilon$ being counted from the ionization threshold, the vertical axis of the diagrams gives the energy of the photoelectrons emitted in the zeroth ATI order. As the basis set used in the calculation is not suitable for resolving the dressed Rydberg states accumulating between −0.25 and 0 eV, modulo $\hbar\omega$, no results are presented between $\hbar\omega-0.25$ eV (1.30 eV) and $\hbar\omega+0$ eV (1.55 eV). Dressed excited-states that do not interact with the dressed ground state under the dipole selection rules are not shown, and neither are the dressed $1s$, $2s$, $2p$, and $3s$ states. Due to numerical instabilities, some of the quasienergy curves could not be followed below or above certain intensities, at which they seem to start or end abruptly.

Being deeply bound, the dressed ground state shifts little in the field, apart from the downward shift by $-U_p$ introduced by the removal of the term proportional to $A^2(t)$ from the Hamiltonian. The solid straight lines indicate where the points corresponding to this state would lie if its Stark shift was exactly $-U_p$. Its actual trajectory (i.e., the trajectory of the dressed $3p_{m=0}$ state of the one-electron model) is traced by the alignments of points closely following the solid lines. All the other quasienergies pertain to dressed excited states. The atom must absorb at least 11 photons to ionize from the ground state in weak fields. As the intensity increases, the ac Stark shift of this state lowers its energy with respect to the continuum limit, with the effect that it passes under the 12-photon ionization threshold at 21 TW cm^{-2}. (That is, 11-photon ionization becomes impossible and 12 photons are required to ionize the atom.) Likewise, the atom passes the 13-photon threshold at 47 TW cm^{-2} and the 14-photon threshold at 73 TW cm^{-2}. Each crossing of an ionization threshold is accompanied by a change in the parity of the Rydberg states the ground state can couple to within the dipole selection rules, and therefore by a change in the set of the dressed excited states the dressed ground states can interact with. The dressed states relevant between the 12- and the 13-photon thresholds are shown in Fig. 1(b), while those relevant below the 12-photon threshold or between the 13- and 14-photon thresholds are shown in Fig. 1(a). The division ensures that the curve crossings visible in these diagrams all correspond to Stark-shift-induced resonances between the corresponding dressed states.

More information about the crossings involving the dressed ground state is provided in Table I. Columns (a) and (b) of the table give the intensity at which each of these occurs and the corresponding photoelectron energy in the zeroth-order ATI peak (i.e., the horizontal and vertical coordinates of these crossings in the diagrams). Column (c) gives the energy width of the resonant states, Γ, at their crossing. It is worth noting that a width of 0.05 eV corresponds to an ionization lifetime $\hbar/\Gamma$ of about 13 fs. Even at large intensities, many dressed excited states have a width of the order of 0.05 eV or smaller at their crossing with the ground state. If populated at the resonance, these states may therefore withstand several optical cycles before being photoionized. Their lifetime may seem surprisingly long, given that all excited states of argon may decay by over-the-barrier ionization at the intensities considered. However, one should keep in mind that over-the-barrier ionization is not efficient if the laser period is shorter than the orbital period (because the electron is then unlikely to have enough speed to leave before the electric field reverts), which is the case for all the excited states important here.

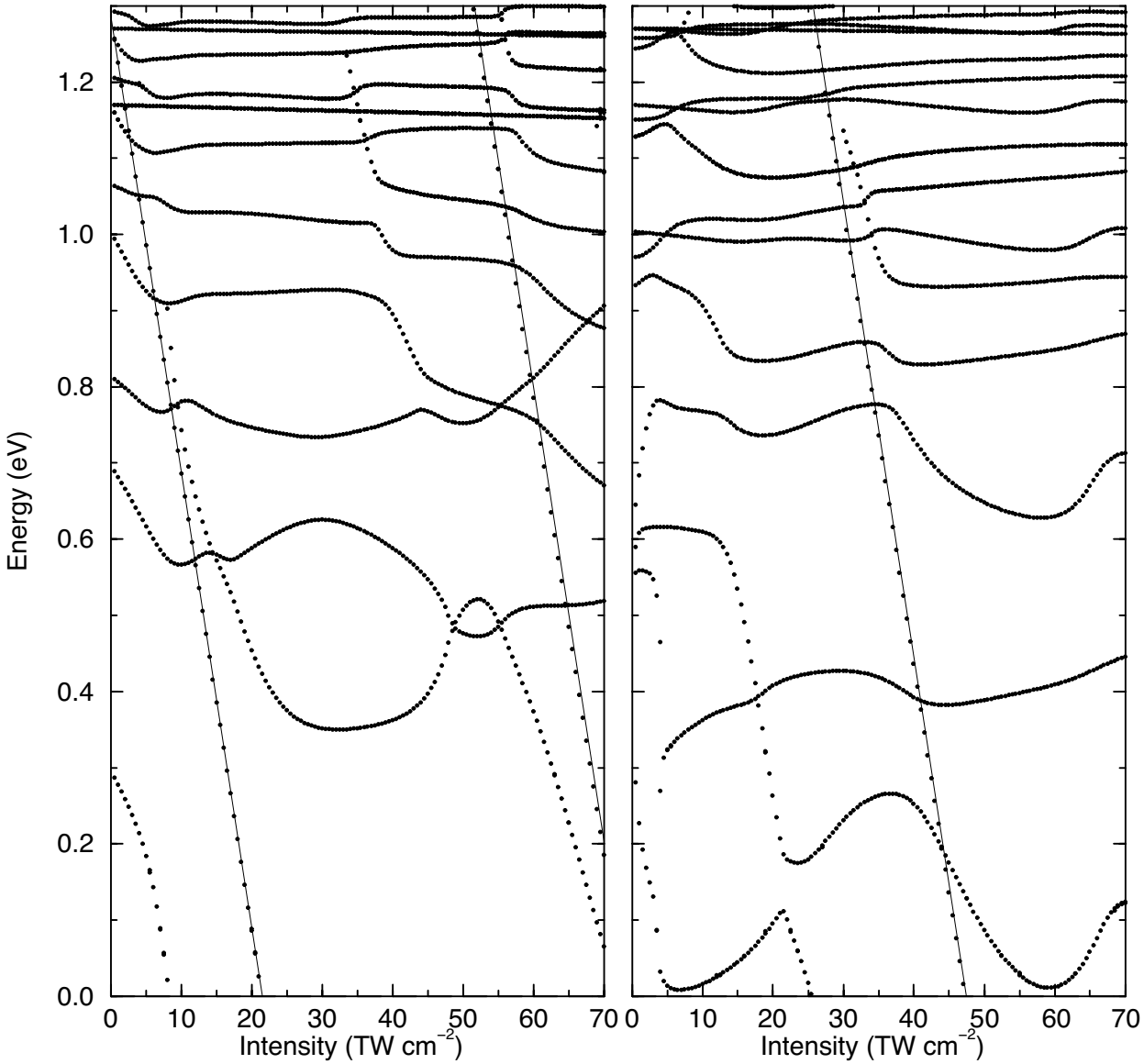

FIG. 1. Real part of the quasienergies of Ar in a laser field of 800 nm wavelength vs intensity. The atom is described by the model potential for the $J=3/2$ core. The solid straight lines represent the trajectory of the dressed ground state in the absence of any nonponderomotive shift. Left-hand side: dressed states interacting with the ground state below 21 TW cm^{-2} and above 47 TW cm^{-2}; right-hand side: dressed states interacting with the ground state between 21 and 47 TW cm^{-2}.

For each crossing, the field-free state that the quasienergy curve of the resonant state reduces to in the zero-field limit is specified in column (d), when known. For convenience, we use these limits as labels identifying the various dressed states. It should be noted, however, that the dressed states often have a different character at high intensity than in weak fields. This can be seen by comparing columns (d) and (e) of the table. The latter identifies the partial waves that dominate the wave function of the resonant state at the crossing intensity [32]. For example, the state developing adiabatically from the $7g$ state has a predominantly s character at its crossing with the ground state at 26 TW cm^{-2}: its wave function is dominated by a partial wave with $l=0$, which accounts for 51% of its norm at that intensity, while its most important partial wave with $l=4$ only contributes 32% of its norm. (We count the photon numbers N with respect to the dominant harmonic component of the dressed ground state. Therefore, the values given in the table also indicate the order of the resonances with the various dressed excited states.) The states of largest angular momentum—the $6h$, $7h$, and $7i$ states—keep their character up to high intensities. These excited states also have a particularly long lifetime and follow straight-line, almost horizontal trajectories in Fig. 1. (The $7i$ state is visible at 1.27 eV in the right-hand side of Fig. 1, although its quasienergy curve is partly hidden by other excited states. The other two states can be seen at about 1.16 and 1.26 eV in the left-hand side of the figure.) Their momentum-space distribution is sharply peaked at $p=0$, which explains why they couple relatively little with the field. Some other states simply swap character in avoided crossings, e.g., the $9s$ and the $7g$, the $8s$ and the $6g$, the $7s$ and the $5g$, and the $8p$ and the $7f$. Low excited states tend to

TABLE I. Details of the crossings visible in Fig. 1 between the dressed ground state and the dressed excited states ($J=3/2$ core). (a) Crossing intensity in TW cm^{-2}. (b) Photoelectron energy at the resonance, within the range $[0, \hbar\omega]$, in eV. (c) Ionization width of the resonant dressed state Γ in eV. (d) Adiabatic zero-field limit of the resonant dressed state. (e) Composition of the resonant state (the photon index N and the orbital angular momentum quantum number l of the dominant partial waves, and their contributions to the norm of the wave function, in %). (f) Same as (b), but for the crossing of the same dressed excited state with the dressed ground state between 73 and 99 TW cm^{-2}.

(a)	(b)	(c)	(d)	(e)	(f)
25.6	1.30	0.014	?	$N=11$, $l=2$ (79%)	
26.0	1.28	0.032	7g	$N=11$, $l=0$ (51%)	1.29
				$N=11$, $l=4$ (32%)	
26.0	1.27	0.016	9s	$N=11$, $l=4$ (46%)	1.27
				$N=11$, $l=0$ (34%)	
26.2	1.27	0.0001	7i	$N=11$, $l=6$ (69%)	1.26
27.0	1.21	0.022	7d	$N=11$, $l=2$ (76%)	1.23
27.5	1.18	0.046	8s	$N=11$, $l=0$ (55%)	1.21
				$N=11$, $l=4$ (23%)	
27.6	1.18	0.034	6g	$N=11$, $l=4$ (45%)	1.17
				$N=11$, $l=0$ (24%)	
29.2	1.08	0.041	6d	$N=11$, $l=2$ (68%)	1.11
30.0	1.04	0.070	7s	$N=11$, $l=0$ (73%)	1.09
30.7	1.00	0.049	5g	$N=11$, $l=4$ (37%)	0.99
				$N=10$, $l=3$ (14%)	
33.0	0.86	0.097	5d	$N=11$, $l=2$ (51%)	0.89
34.3	0.78	0.140	6s	$N=11$, $l=0$ (58%)	0.69
40.8	0.39	0.231	?	$N=11$, $l=0$ (29%)	0.50
				$N=11$, $l=2$ (15%)	
44.1	0.19	0.360	4d	$N=10$, $l=1$ (15%)	0.01
				$N=11$, $l=0$ (10%)	
51.7	1.29	0.029	9p	$N=12$, $l=3$ (68%)	
52.0	1.26	0.005	7h	$N=12$, $l=5$ (53%)	
52.2	1.25	0.038	7f	$N=12$, $l=1$ (64%)	
53.2	1.19	0.048	8p	$N=12$, $l=3$ (58%)	
53.8	1.16	0.005	6h	$N=12$, $l=5$ (34%)	
				$N=11$, $l=4$ (15%)	
				$N=13$, $l=6$ (11%)	
54.1	1.14	0.062	6f	$N=12$, $l=1$ (56%)	
55.8	1.04	0.086	?	$N=12$, $l=3$ (40%)	
57.1	0.96	0.134	7p	$N=12$, $l=1$ (33%)	
				$N=11$, $l=0$ (11%)	
59.6	0.81	0.400	6p	$N=11$, $l=0$ (18%)	
				$N=11$, $l=2$ (17%)	
				$N=12$, $l=1$ (17%)	
60.5	0.75	0.203	5f	$N=12$, $l=3$ (13%)	
				$N=11$, $l=2$ (12%)	
64.5	0.51	0.321	4f	$N=12$, $l=1$ (25%)	
				$N=10$, $l=1$ (11%)	

lose their zero-field identity even at moderate intensities. For example, the dressed 6p state has a mixed p and f character at its first crossing with the ground states, at 8.6 TW cm^{-2}, and no clear character at its second crossing, at 59.6 TW cm^{-2}. The dressed 4f state, which is dominated by $l=1$ partial waves when it crosses the ground state at 64.5 TW cm^{-2}, has already acquired a p character at the relatively low intensity of 10 TW cm^{-2}.

The dressed ground state is much more stable than the dressed excited states considered here. Its energy width is at most 5.5×10^{-5} eV below 70 TW cm^{-2}, corresponding to a lifetime of at least 4500 optical cycles. Because of this discrepancy in energy widths, one can expect that all the crossings it undergoes are direct in the real part of the quasienergy [22,23]. This is borne out by the absence in Fig. 1 of avoided crossings involving the ground state. However, we cannot exclude that some of the crossings are actually avoided rather than direct, with an unresolved energy gap of the order of 5 meV or less. Physically, there is little difference between these two cases, because the variation of the intensity in a realistic laser pulse would be fast enough for the atom to jump diabatically across any energy gap of that width. Given the speed at which the ground state shifts with respect to the excited states, the two dressed states involved would then interchange character within a negligibly small region of intensity. Whether their crossing is avoided or not, the two states may influence each other over the whole range of intensity where they overlap in energy, with the dressed ground state acquiring some of the character of the resonant state and conversely. The resonances may thus affect ionization from the ground state without transfer of population to the dressed resonant state [33].

The changes in the wave function of the dressed ground state between 54 and 70 TW cm^{-2} are illustrated by Fig. 2. The resonant states are dominated by their $N=12$ harmonic component in this range of intensity, with the exception of the dressed 6p state. Correspondingly, certain partial waves of the $N=12$ harmonic component of the dressed ground state are enhanced as it shifts through the width of the resonant states. Note, in particular, the large increase of the f wave around 60 TW cm^{-2} and of the p wave around 64 TW cm^{-2}. These increases are concomitant with the crossings, respectively, of the dressed 5f state (which has a dominant f character) and of the dressed 4f state (which has a dominant p character). The width of the ground state and the shift of the resonant states being both negligible, the intensity width of each enhancement reflects the energy width of the resonant state. Since ionization from the dressed ground state may be enhanced in the whole range of intensity where its quasienergy falls within the width of the resonant state, the width of the corresponding resonance peak in the ATI spectrum will be equal to the ionization width of the dressed excited state, even without transfer of population to the latter.

B. Resonances in the ATI energy spectra

We now examine the ATI energy spectra obtained at 800 nm by Nandor *et al.* [6]. The pulses used in their

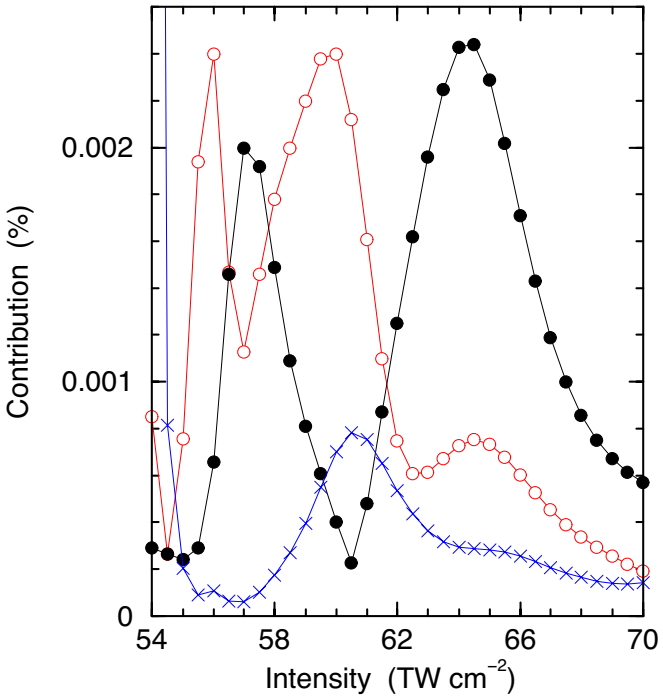

FIG. 2. (Color online) Contribution of the $l=1$ (closed circles), 3 (open circles), and 5 (crosses) partial waves of the $N=12$ harmonic component of the dressed ground state to the norm of its wave function, in %. The range of intensity considered covers the last six entries of Table I.

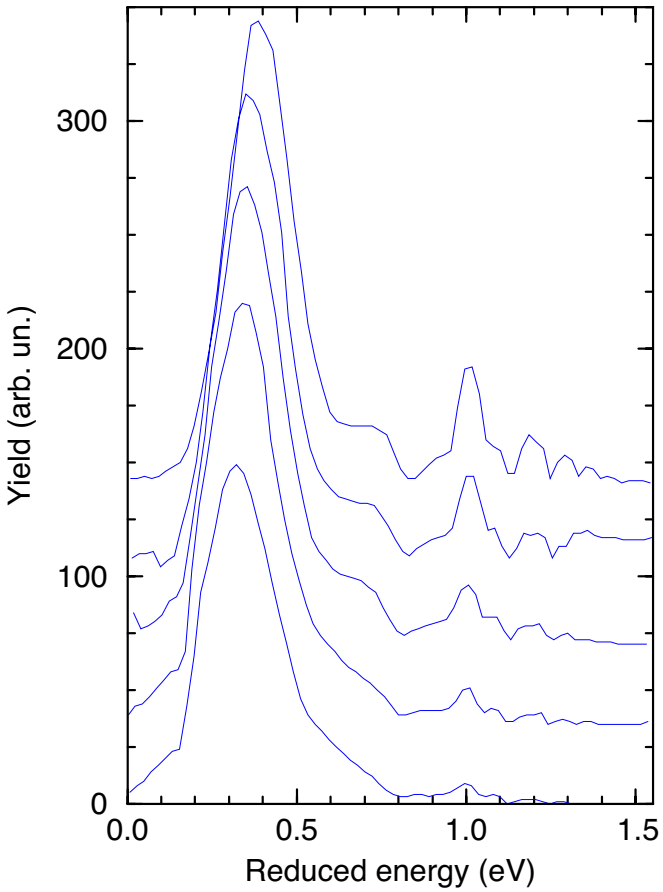

FIG. 3. (Color online) Theoretical ATI energy spectrum for an experimental 800 nm pulse of 44.5 TW cm^{-2} peak intensity, as calculated by numerically integrating the time-dependent Schrödinger equation [6]. Solid curves, from top to bottom: yield in the 12th, 13th, 14th, 15th, and 16th ATI orders. For clarity, each curve has been shifted vertically by an arbitrary amount. The horizontal axis is the photoelectron energy reduced to the interval $[0, \hbar\omega]$. Adapted from Ref. [35], with permission.

experiment had a duration of 120 fs. Their calculations were done for 45-cycle flat-top pulses smoothly turned on over an half optical cycle. An ATI spectrum was calculated for a number of such pulses. These results were then averaged over the spatial and temporal distributions of intensity of the experimental pulses and over the two core symmetries of the excited states, so as to yield theoretical spectra that could be compared to the data. We concentrate here on the theoretical spectra, as they reproduce all the features of the experimental spectra but have a higher resolution. The photoelectron yield is shown in Figs. 3–5 below, for three different pulse peak intensities and for five consecutive ATI orders belonging to the plateau part of the spectrum. For each order, the yield is plotted as a function of the reduced energy, $E_r = E_{ph} - S\hbar\omega$, where E_{ph} is the photoelectron energy and S is the number of excess photons ($S=1$ in the first order, 2 in the second order, etc.) Photoelectron energies differing by integral multiples of $\hbar\omega$ all correspond to the same value of E_r, and E_r varies between 0 and $\hbar\omega$.

The agreement between calculated and measured spectra shows that the plateau resonances can be understood from the response of the atom to a stationary field. It is therefore meaningful to compare the results of Figs. 3–5 to the quasienergy spectrum of Fig. 1. However, since most of the relevant resonances overlap to some extent, the structures they induce in the yield may vary from ATI order to ATI order and may be more complex than simple Lorentzian peaks. The profile of the resonances may also be affected by the finite resolution of the calculation and by the averaging over the distribution of intensity. In some cases, it may also be slightly broadened by the difference in energy between the two core symmetries.

Transfer of population between dressed states is possible when the intensity varies [34]. We expect that the fast turnon of the flat-top pulses used in the calculations of Ref. [6] left the atom in a superposition of the dressed ground state (mainly) and of the neighboring resonant states (to a small extent). A resonance structure in the ATI spectrum may thus originate primarily from ionization from one or several of these dressed excited states. Alternatively, if there was little or no transfer, it may originate primarily from ionization from the dressed ground state. The Floquet map alone does not offer much information about which of these two mechanisms might predominate. However, it is likely that both produce similar resonance structures, since, as we have seen at the end of last section, dressed states imprint their "image" in the dressed ground state.

1. Low intensities

Let us first consider ionization in a pulse with a peak intensity of 44.5 TW cm^{-2} (Fig. 3). This field is strong

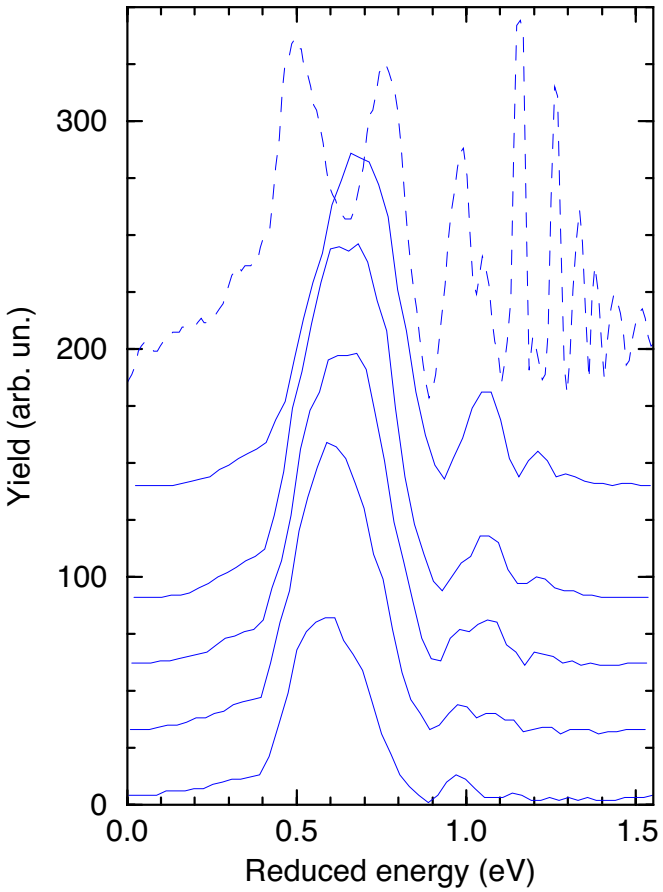

FIG. 4. (Color online) As in Fig. 3. Here, the peak intensity of the pulse is 66.1 TW cm^{-2}. The dashed curve shows the yield in the first ATI order (not to scale) [6].

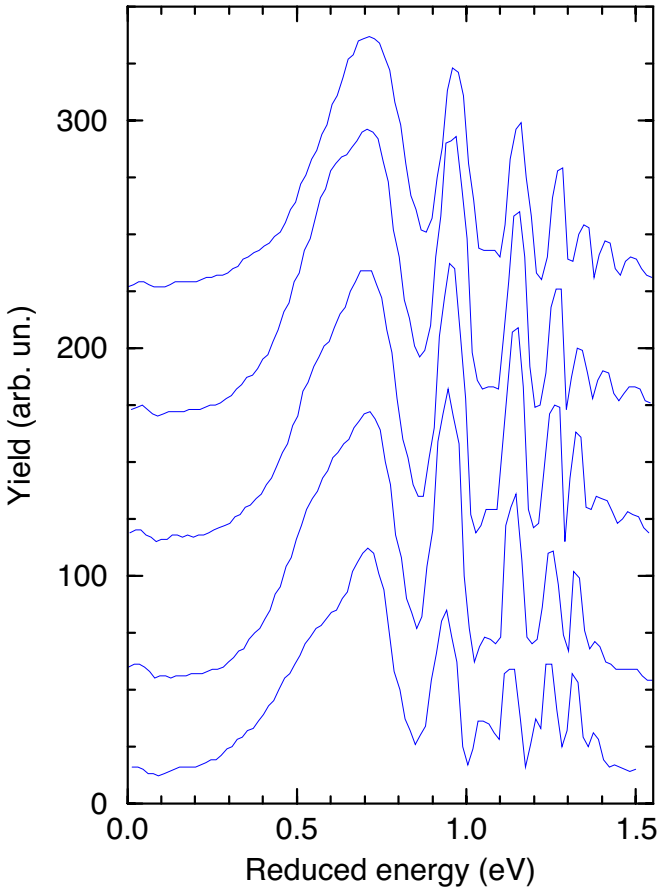

FIG. 5. (Color online) As in Fig. 3. Here, the peak intensity of the pulse is 94.2 TW cm^{-2}.

enough that the ground state may be in resonance with the dressed excited states shown in the right-hand side of Fig. 1. (The states shown in the left-hand side can be ignored here as emission in the 12th and higher ATI orders is negligible below 21 TW cm^{-2}.) The details of the relevant crossings are provided in the top half of Table I.

The two most prominent features of the low-order ATI spectrum in such relatively weak pulses are a narrow peak at $E_r=1$ eV and a broad enhancement covering much of the region below 0.7 eV [8,13]. (Here and in the following, all energies refer to reduced energies E_r. The structures mentioned are visible in several ATI orders.) The analysis of the time-dependent wave function shows that the narrow peak originates from a resonance with the $5g$ state [8,13]. The same conclusion can be drawn from the quasienergy spectrum, as the maximum is centered at the energy expected for electrons detached at the crossing of the dressed ground state with the dressed $5g$ state. This peak is repeated at the same position in all ATI orders but is less prominent in the high-energy part of the spectrum. It is still well marked in the 12th order and it gradually disappears in the higher orders. The time-dependent calculations indicate that recollisional ATI is unlikely for the $5g$ state at the intensities involved [8]. This peak is best seen as a simple case of high-order, resonantly enhanced multiphoton absorption, similar in mechanism to the familiar resonance enhancements of low-order ATI.

More resonance structures are visible above $E_r=1$ eV, particularly in $S=12$ and 13. (The higher ATI orders fall well beyond the cutoff of the energy distribution.) Most of these structures can be referred to particular curve crossings, using the data provided in Table I. Like the $5g$ peak, they probably arise from nonrecollisional multiphoton absorption.

The broad enhancement below $E_r=0.7$ eV in the low-order spectrum has been assigned to a double resonance with the $4p$ and $4d$ states accompanied by nonresonant ionization [13,36]. It certainly occurs in a region of intensity where the ground state interacts with two broad, overlapping dressed excited states (Fig. 1, Table I). One, of unknown origin crosses the dressed ground state at 0.39 eV in the $J=3/2$ spectrum and at 0.47 eV in the $J=1/2$ spectrum. The other originates from the $4d$ state and crosses at, respectively, 0.19 and 0.29 eV. These two states participate in a sequence of avoided crossings with large energy gaps, which might further complicate the ground state's ionization dynamics. Like the first few ATI orders, the plateau part of the spectrum is also marked by a conspicuous resonance peak in this range of reduced energies. For $S=12$, the maximum of this peak coincides within 0.01 eV with the reduced energy expected for electrons detached at the crossing with the state of unknown origin mentioned above. The width of this peak is also consistent with the ionization width of this state. The maximum shifts to smaller energies for $S>12$ but remains within a range consistent with an enhancement primarily due

to this dressed state. Given the width of this peak, the dressed $4d$ state and nonresonant ionization might also contribute. Resonance with the $4d$ state seems to modulate the low-energy wing of this peak, around and below 0.2 eV, by a broad enhancement for $S=15$ and 16 and by a crest-and-trough Fano profile for $S=13$ and 14. There is clear evidence for nonresonant ionization in the spectrum of the photoelectrons ejected at 90° from the polarization direction [12]. However, the importance of nonresonant ionization in the direction of polarization is difficult to ascertain.

2. Middle intensities

The ATI spectrum at a peak intensity of 66.1 TW cm^{-2} is presented in Fig. 4, for the same ATI orders as in Fig. 3 and also for $S=1$ (the dashed curve). Ionization is sufficiently stronger above 50 TW cm^{-2} that the resonance enhancements occurring at lower intensities are almost completely masked by new enhancements occurring between 50 and 66 TW cm^{-2}. The former only manifest by the barely visible shoulders at $E_r \approx 0.35$ eV, which for $S \geq 12$ are the remnants of the large peaks so conspicuous in Fig. 3 [37]. The atom needs to absorb at least 13 photons to lose an electron from the ground state, here, and the relevant dressed states are now those represented in the left-hand side of Fig. 1. The details of the crossings are given in the bottom half of Table I.

While the low-order spectrum exhibits a number of narrow resonances up to close to $E_r = \hbar\omega$, the high-order spectrum is dominated by a large enhancement, between 0.4 and 0.8 eV, and by a few smaller peaks between 0.8 and 1.3 eV. The large enhancement has its maximum a little below 0.7 eV in $S=12$, which is not inconsistent with a resonance with the dressed $5f$ state. However, the maximum occurs at lower energies in higher ATI orders and tends towards the position of the crossing with the dressed $4f$ state. By contrast, the resonances with the dressed $4f$ and $5f$ states are both individually visible in the first ATI order. This double peak for $S=1$ evolves continuously into a single peak in higher ATI orders, as can be seen from the full ATI spectra of Ref. [6]. We therefore believe that these enhancements originate from these two Stark-shift-induced resonances, in the low orders as well as in the high orders. Its significant drift between the 12th and the 16th orders may seem difficult to reconcile with a picture of ionization from dressed excited states populated during the turnon of the pulse. However, the electrons contributing to this enhancement are almost certainly emitted from the dressed ground state, not from dressed excited states. Indeed, Muller and Kooiman [7], when looking at the quasistationary state producing the plateau electrons at intensities between the $4f$ and the $5f$ resonances, noted that this state was decreasing, due to ionization, by three parts in 10^4 at each optical cycle in the flat part of the trapezoidal pulses used in their calculations. This decrease matches that of the dressed ground state and is four orders of magnitude slower than that of the two resonant states. We have seen above that the dressed ground state is perturbed by the influence of the dressed $4f$ and $5f$ states between the crossings as well as at the crossings. From this, we surmise that the emission of plateau electrons can be resonantly enhanced even at energies differing somewhat from those of the crossings. However, the recolliding wave packets must also interfere constructively (with themselves and with the rest of the wave function) for this to happen [8]. The interference is likely to limit the intervals of intensities (hence, of photoelectron energies) over which strong emission takes place. As these intervals can be expected to be slightly different in different ATI orders, there is no contradiction between the possibility of resonantly enhanced ionization and the change in the position of the "$4f$-$5f$" peak. The importance of population transfer to the resonant dressed states depends on the pulse shape. It may be larger for pulses used in actual experiments than for the flat-top pulses used in the calculations. Nevertheless, given the excellent agreement between the calculated and the measured spectra, we believe that population transfer does not play a significant role for this particular plateau resonance in the experiments of Nandor *et al.* [6].

The requirement of a constructive interference might also make a same Stark shift induced resonances enhancing ionization in certain ATI orders but not in others. An example of such selective enhancements is found in Fig. 4. The two peaks around 1.05 and 1.21 eV in the 12th and 13th orders are consistent with resonances with a dressed state of unknown origin, which crosses the ground state at 1.04 eV, and with the dressed $8p$ state, which crosses at 1.19 eV. However, these two peaks decrease in importance for $S>12$, at the same time as two new ones grow at 0.96 and 1.14 eV. Only the latter two show up in the 16th order. (We tentatively identify these as resonances with the dressed $7p$ and $6f$ states, since their positions coincide with the corresponding crossing energies. As is noted in the Appendix, where the $S=1$ spectrum is examined, three of these four crossings are also clearly manifest in the low-order spectrum.)

3. High intensities

Part of the ATI plateau at 94.2 TW cm^{-2} peak intensity is shown in Fig. 5. The electrons detached between 60 and 70 TW cm^{-2} are responsible for much of the large enhancement below $E_r=0.85$ eV. Far more are detached above 70 TW cm^{-2} than below 60 TW cm^{-2}, with the result that all the peaks above $E_r=0.85$ eV are new resonances. Apart from the exception mentioned below, the relevant dressed states are the same as in the case of Fig. 3. We have calculated the quasienergy of the states shown in the right-hand side of Fig. 1 beyond 70 TW cm^{-2}, up to their next crossing with the dressed ground state. The photoelectron energy at each high-intensity crossing is given in column (f) of Table I. The crossing intensities are higher by about 52 TW cm^{-2} than those specified in column (a) of this table.

Comparing the spectra at intensities intermediate between those of Fig. 4 and Fig. 5 shows that the faint resonance profile visible below 0.2 eV in the latter is due to ionization between 70 and 73 TW cm^{-2} (for $J=3/2$). The ground state is within the width of a broad excited state at these intensities. Although they do not cross each other, the proximity of this resonance makes it likely that it influences ionization from the ground state and produces a structure in the high-order spectrum.

The two new significant enhancements below 1 eV seem to be straightforward Stark-shift-induced resonances. One, peaking at 0.70 eV, is superimposed on the $4f$-$5f$ peak coming from ionization at middle intensities; the other peaks at 0.95 eV. These two new resonance change little in position between $S=12$ and 16. In view of their position, they can be ascribed, respectively, to the dressed $6s$ state and to a state of unknown origin which crosses the ground state at 0.96 eV. (This state appears at 30 TW cm^{-2}, in the right-hand side of Fig. 1, at too high an intensity to cross the ground state below 70 TW cm^{-2}. It is not mentioned in Table I.) Surprisingly perhaps, the dressed excited state that produces the large peak at $E_r \approx 0.4$ eV in Fig. 3 does not give rise to any enhancement in Fig. 5.

The spectrum is more complex above 1 eV. This region corresponds to intensities where the ground state crosses a number of overlapping dressed excited states. The spectrum changes significantly between $S=12$ and 16, with some of the peaks shifting appreciably and new maxima developing between them. These structures are difficult to assign to resonances with particular dressed states in the absence of any other information than their position. As in the case of the $4f$-$5f$ resonance at lower intensities, it is possible that several resonant states are capable of enhancing the emission of hot electrons in this region.

It is striking, if we compare Figs. 3–5, that the leftmost major enhancement occurs at higher and higher reduced energies as the intensity increases. This implies that the lowest resonant dressed excited states responsible for the emission of hot electrons are higher and higher in the quasienergy spectrum. This shift possibly originates in the growing amplitude of the quiver motion of the electron, which enables electrons initially more and more distant to recollide with the core. The recollision may involve electrons initially in a dressed excited state; however, this does not need to be the case since resonant states mix with the dressed ground state over the whole interval of intensity where they overlap in energy.

IV. CONCLUSIONS

In conclusion, we have calculated the quasienergy spectrum of argon submitted to an intense 800 nm laser field and have related the plateau resonances to the Stark-shift-induced resonances predicted by this calculation.

The quasienergy spectrum is complex, and a simple picture of states shifting linearly as the intensity increases is incorrect for most of the excited states of interest. Many of them have multiple avoided crossings with each other and only a few can be identified with field-free excited states up to high intensity. However, the ionization width of many of the important dressed excited states is narrow enough to explain the sharp enhancements of high-order ATI observed for pulses of long duration.

Our results support the interpretation of these enhancements as arising from Stark-shift-induced resonances. They occur at intensities where resonant excited states mix with the ground state in the dressed-ground-state wave function. Often, but not always, they coincide with a crossing between the corresponding quasienergy curves. We have found no evidence for enhancements that are not concomitant with a perturbation of the dressed ground state by one or several dressed excited states. Such enhancements would likely be channel-closing enhancements rather than Stark-shift-induced resonances.

In many cases, a same resonant state seems responsible for an enhancement of ATI in the low orders as well as in the high orders. Because of the mixing, ionization from the ground state can be resonantly enhanced without population present in a dressed excited state. In particular, there is strong evidence that the most intense plateau enhancement below 70 TW cm^{-2}, the $4f$-$5f$ peak, arises from ionization from the dressed ground state, not from ionization from a dressed excited state.

Below 40 TW cm^{-2}, there seems to be little difference between resonances in the low ATI orders and resonances in the high ATI orders, apart from the magnitude of the enhancement. At these relatively low intensities, emission of hot electrons can be ascribed to nonrecollisional multiphoton absorption. Recollision is clearly important at higher intensities [7,8], and it seems that Stark-shift-induced resonances often create conditions favorable for this process. Overlapping resonances with similar potential for recollision are likely to interfere with each other, which may explain why there is no one-to-one correspondence between plateau enhancements and quasienergy curve crossings in certain regions of intensity where the ground state sweeps through multiple resonances. However, we still find sharp enhancements associated with single, identifiable dressed excited states well above 80 TW cm^{-2}. Some of the resonant dressed states of interest are short lived at the relevant intensities—for example, the dressed $4f$ and $5f$ states both have a lifetime of less than two optical cycles between 60 and 65 TW cm^{-2}. Nonetheless, they affect the dressed ground state, as shown by Fig. 2.

The analyses in Refs. [7,8,10] of the time-dependent wave function reveals that at high intensity a remarkable bunching of wave packets starts when part of the electronic density is trapped in the Coulomb tail of the atomic potential, at about 3 a.u. from the nucleus. Furthermore, at intensities at which the emission in the high ATI orders is particularly intense, the wave function has maxima close to the nucleus, with near-zero drift velocity, which produce bursts of high-energy electrons upon backscattering on the nucleus. (For this to happen, the resonant state should also be relatively low in the spectrum and, at least at the intensities considered here, should have an orbital angular momentum not larger than 3—see Table I.) As will be reported elsewhere, this increase of electronic density close to the nucleus also manifests in the Floquet wave function, particularly in the partial wave of lowest l value of the harmonic component corresponding to the order of the highest ATI peak in the plateau: The mean radius of this partial wave shrinks to values below 1 a.u.. and passes through a broad minimum at the intensities where the emission of high-energy electrons has been observed [6]. The part of the electronic wave packet described by this partial wave is thus located close to the nucleus, where it can absorb many photons from the laser field.

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APPENDIX: RESONANCES IN LOW ATI ORDERS

In this appendix, we identify the Stark-shift-induced resonances manifesting in the low ATI orders at 66.1 TW cm^{-2} peak intensity. The photoelectron spectrum is represented by a dashed curve in Fig. 4.

In view of their position, and in agreement with the wave-function analysis [8], we interpret the tall peaks at 1.16 and 1.26 eV as due to electrons ionized at resonance with, respectively, the dressed $6h$ and the dressed $7h$ states. Since the ionization widths of the h series are much smaller than the widths of the corresponding ATI peaks, it is likely that neighboring resonances with the dressed $6f$ and $7f$ states also contribute to these peaks. We identify the small peak at 1.20 eV to the dressed $8p$ state, since it occurs at an energy almost exactly equal to that expected for this resonance. The doublet with subpeaks at 1.00 and 1.05 eV can be associated with the two crossings at 0.96 and 1.04 eV, respectively with a state of unknown zero-field limit but clear f character at the resonance, and with the dressed $7p$ state. The crossing with the dressed $6p$ state does not seem to induce any structure in the low-energy spectrum. The broad peak centered at 0.50 eV develops on a background originating from ionization below 45 TW cm^{-2}. This peak and the one centered at 0.76 eV have the energies and the widths expected for resonances with, respectively, the dressed $4f$ state (which has a p character at the resonance) and the dressed $5f$ state (which has f character at the resonance). The character of these resonant states is echoed by the larger importance at the corresponding crossings of the p and f partial waves of the $n=12$ harmonic component of the dressed ground state (Fig. 2). It is also consistent with the time-dependent wave function of Refs. [8,13] having an off-axis component of p symmetry in the high-intensity wing of this double hump and of f symmetry in the low-intensity wing.

[1] See, e.g., W. Becker, F. Grasbon, R. Kopold, D. B. Milošević, G. G. Paulus, and H. Walther, Adv. At., Mol., Opt. Phys. **48**, 35 (2002).

[2] B. B. van Linden van den Heuvell and H. G. Muller, in *Multiphoton Processes*, edited by S. J. Smith and P. L. Knight (Cambridge University Press, Cambridge, U.K., 1988), p. 25; P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993); K. C. Kulander, K. J. Schafer, and J. L. Krause, in *Super-Intense Laser-Atom Physics*, edited by B. Piraux, A. L'Huillier, and K. Rzążewski (Plenum Press, New York, 1993), p. 95.

[3] R. R. Freeman, P. H. Bucksbaum, H. Milchberg, S. Darack, D. Schumacher, and M. E. Geusic, Phys. Rev. Lett. **59**, 1092 (1987).

[4] P. Hansch, M. A. Walker, and L. D. Van Woerkom, Phys. Rev. A **55**, R2535 (1997).

[5] M. P. Hertlein, P. H. Bucksbaum, and H. G. Muller, J. Phys. B **30**, L197 (1997).

[6] M. J. Nandor, M. A. Walker, L. D. Van Woerkom, and H. G. Muller, Phys. Rev. A **60**, R1771 (1999).

[7] H. G. Muller and F. C. Kooiman, Phys. Rev. Lett. **81**, 1207 (1998).

[8] H. G. Muller, Phys. Rev. A **60**, 1341 (1999).

[9] H. G. Muller, Phys. Rev. Lett. **83**, 3158 (1999).

[10] H. G. Muller, Opt. Express **8**, 44 (2001).

[11] H. G. Muller, Opt. Express **8**, 86 (2001).

[12] E. Cormier, D. Garzella, P. Breger, P. Agostini, G. Chériaux, and C. Leblanc, J. Phys. B **34**, L9 (2001).

[13] R. Wiehle, B. Witzel, H. Helm, and E. Cormier, Phys. Rev. A **67**, 063405 (2003).

[14] G. G. Paulus, F. Grasbon, H. Walther, R. Kopold, and W. Becker, Phys. Rev. A **64**, 021401(R) (2001); R. Kopold, W. Becker, M. Kleber, and G. G. Paulus, J. Phys. B **35**, 217 (2002).

[15] B. Borca, M. V. Frolov, N. L. Manakov, and A. F. Starace, Phys. Rev. Lett. **88**, 193001 (2002); S. V. Popruzhenko, Ph. A. Korneev, S. P. Goreslavski, and W. Becker, *ibid.* **89** 023001 (2002).

[16] J. Wassaf, V. Véniard, R. Taïeb, and A. Maquet, Phys. Rev. Lett. **90**, 013003 (2003); Phys. Rev. A **67**, 053405 (2003).

[17] C. C. Chirilă and R. M. Potvliege, Phys. Rev. A **71**, 021402(R) (2005); F. H. M. Faisal and G. Schlegel, J. Phys. B **38**, L223 (2005); D. G. Arbó, S. Yoshida, E. Persson, K. I. Dimitriou, and J. Burgdorfer, Phys. Rev. Lett. **96**, 143003 (2006).

[18] F. Grasbon, G. G. Paulus, H. Walther, P. Villoresi, G. Sansone, S. Stagira, M. Nisoli, and S. De Silvestri, Phys. Rev. Lett. **91**, 173003 (2003).

[19] M. Wickenhauser, X. M. Tong, and C. D. Lin, Phys. Rev. A **73**, 011401(R) (2006).

[20] J. H. Shirley, Phys. Rev. **138**, B979 (1965); Shih-I Chu, Adv. At. Mol. Phys. **21**, 197 (1985).

[21] Y. Gontier and M. Trahin, Phys. Rev. A **7**, 1899 (1973).

[22] C. R. Holt, M. G. Raymer, and W. P. Reinhardt, Phys. Rev. A **27**, 2971 (1983).

[23] R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **40**, 3061 (1989).

[24] R. M. Potvliege and R. Shakeshaft, Adv. At., Mol., Opt. Phys. **1(Suppl)**, 373 (1992).

[25] A. Maquet, Shih-I Chu, and W. P. Reinhardt, Phys. Rev. A **27**, 2946 (1983).

[26] R. M. Potvliege, Comput. Phys. Commun. **114**, 42 (1998).

[27] S. Vučić, Phys. Rev. A **65**, 033421 (2002).

[28] The quasienergy spectrum of argon at 248 nm has been calculated by Plummer and Noble using the R-matrix–Floquet method [M. Plummer and C. J. Noble, J. Phys. B **33**, L807 (2000); **34**, L51 (2002); **36**, L219 (2003)].

[29] The difference is less than 0.01 eV for all the excited levels above the $5s$, and 0.01–0.02 eV for the $4p$, $3d$, and $5s$ states. In the context of a fully time-dependent calculation, it is ad-

vantageous to remove the 1*s*, 2*s*, and 2*p* states by adjusting the short-range part of the model potentials [7,8]. No adjustment is necessary here, as these states are too weakly coupled to the dressed states of interest.

[30] C. J. Joachain, M. Dörr, and N. J. Kylstra, Adv. At., Mol., Opt. Phys. **42**, 225 (2000).

[31] R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 6190 (1988).

[32] We define the dominant partial waves as being those that contribute at least 10% of the norm of the wave function, the norm being calculated as the cycle-average overlap of the wave function with the complex conjugate of its dual.

[33] H. Rottke, B. Wolff-Rottke, D. Feldman, K. H. Welge, M. Dörr, R. M. Potvliege, and R. Shakeshaft, Phys. Rev. A **49**, 4837 (1994).

[34] H. C. Day, B. Piraux, and R. M. Potvliege, Phys. Rev. A **61**, 031402(R) (2000).

[35] L. D. Van Woerkom *et al.*, www.physics.ohio-state.edu/~lvw/what/singlee/singlee.html; see also L. D. Van Woerkom, M. A. Walker, M. J. Nandor, and H. G. Muller, in *Multiphoton Processes*, edited by L. F. DiMauro, R. R. Freeman, and K. C. Kulander (AIP, Melville, NY, 2000), p. 70.

[36] However, we find that the dressed 4*p* state cannot be followed as a continuous quasienergy curve above 5 TW cm^{-2}.

[37] These peaks are more visible at slightly lower intensities. Together with the two largest peaks visible in Fig. 4, they form the "subpeak triplets" observed by Hertlein, Bucksbaum, and Muller in each ATI order of the plateau [5].