Constructive and destructive interferences of Stark resonances induced by an ac field in atomic hydrogen

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We study theoretically the problem of a hydrogen atom exposed both to a static dc field and to a monochromatic ac field. We show that, in the presence of an ac field, a constructive (or destructive) interference occurs between the excited (Rydberg) Stark resonance states and the hydrogenic ground state. This mechanism is responsible for dramatic enhancement (or suppression) of the corresponding photoionization rates.

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Strong laser physics is currently one of the most active fields in theoretical and experimental atomic physics. See, for example, Ref. [1] and references therein. Recent technological developments enable one to carry out experiments that were not possible before. See, for example, the most recent study of femtosecond electronic response of atoms to ultraintense x rays [2], and the investigation of acceleration of neutral atoms in strong short-pulse laser fields [3]. These kinds of studies have become feasible because neutral atoms can survive a strong laser field in (long-lived) excited states [4]. From a theoretical point of view, the corresponding experimental high-harmonic generation spectra can be calculated using a single long-lived atomic resonance state [5]. However, even the relatively long-lived resonance atomic states decay fast enough to reduce substantially the harmonic generation yields (see Ref. [1] and references therein, and a review in Refs. [6] and [7]).

In this paper, we address the effect of a dc field on the photoionization (PI) rate of atomic hydrogen. As we show in the following, an effect of the constructive and destructive interferences between the different Stark resonances of atomic hydrogen on the efficiency of the PI process is large, even under small variations of the dc field strength. The situation is completely different for helium atoms, where, for the ground state, the dc field has virtually no effect on the PI decay rate induced by the strong ac field [8]. Ivanov and Kheifets pointed out with some caution that the PI resonance width of helium can be decreased with the dc field [8]. The possibility of suppressing the PI induced by the ac field by an additional weak dc field was first noted by Cocke and Reichl [9] for a δ-function potential and for a selected range of frequencies of the incident laser field. More recently, it has been shown that the suppression of the photoinduced decay rate can occur in a broad and continuous range of the laser parameters [10]. However, a simple explanation was still missing for the PI suppression (and also for dramatic enhancement) of the efficiency of the PI process by varying the weak dc field. This aspect of our understanding of the dramatic effect of a weak dc field on the photoinduced dynamics of atoms in strong ac fields represents the main focus of this work.

Before describing our analysis and results, we should mention that, since one of the crucial factors for improving the efficiency of many light-induced processes is to lower the PI rate, the suppression of PI has been the focus of numerous theoretical and experimental investigations (see Refs. [11,12] and references therein). Among the various stabilization mechanisms, let us mention the Kramers-Henneberger stabilization (KHS) [13] and interference stabilization (IS) [14]. The KHS requires the field intensity to be sufficiently strong and the field frequency sufficiently high such that the quiver-motion amplitude is much larger than the extent of the field-free potential. The interference stabilization, on the other hand, requires an interaction of several Rydberg bound states using two-color lasers. We emphasize this point because, in our studies presented in this paper, the suppression of the PI is achieved as a result of the interaction of different Stark-Rydberg resonance states, which are introduced by applying a one-color laser field. Physical evidence suggests that the second strong laser in Fedorov studies [12,14] may possibly play a role analogous to a weak dc field in our setup.

Let us now describe our theoretical analysis of the resonances of atomic hydrogen in combined ac and dc fields. The corresponding resonance energies and wave functions have been calculated by using the complex scaling method [15]. For a review on the use of complex scaling for calculating resonances of atoms in ac and dc fields, see Chu [16]. In the case of atomic hydrogen, the sought-after resonances are obtained as eigenstates of the Floquet operator

$$\hat{\mathcal{H}} = -i\hbar\partial_t - \frac{\hbar^2}{2m_e}\partial_r^2 + \frac{\hat{L}^2(\theta,\phi)}{2m_e r^2} - \frac{e^2}{r} + er\cos\theta[\epsilon_{\rm dc} + \epsilon_{\rm ac}\cos(\omega t)], \qquad (1)$$

where e and m_e are, respectively, the charge and the mass of the electron, $\hat{L}^2$ is the squared orbital angular momentum, and θ represents an angle between the position vector of the electron and the dc and/or ac electric-field vectors. Note that the ac field is linearly polarized along the z direction. The radial coordinate r is rotated into the upper half of the complex plane so as to make the resonance wave functions square integrable, which

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is similar to bound states in the conventional (i.e., Hermitian) formalism of quantum mechanics [17,18].

The complex-scaled Floquet eigenvalue problem

$$\hat{\mathcal{H}}\phi_n(r,\theta,t) = \left(E_n - \frac{i}{2}\Gamma_n\right)\phi_n(r,\theta,t) \tag{2}$$

is solved using the usual Fourier expansion

$$\phi_n(r,\theta,t) = \sum_m e^{i\omega m t}\varphi_{m,n}(r,\theta). \tag{3}$$

In our numerical calculations, we have used 11 Floquet channels (i.e., $|m| \leqslant 5$). The wave-function components $\varphi_{m,n}(r,\theta)$ are represented in a basis set consisting of 420 Slater-type orbitals (STO) of $L = 0,\ldots,5$ and $M = 0$. For each value of L, we have taken 70 STOs (see Ref. [19] for more details). Such a basis set has proved to yield converged results.

In all of our computations, the ac field frequency was $\omega = 0.633$ a.u. (i.e., 2.62×10^{16} Hz); the atomic units used here are the Hartree energy units. Therefore, the frequency value yields directly the photon energy in atomic units. This value corresponds to the excitation of the hydrogen atom from the ground state to the energy position in the continuum at 0.133 a.u. (3.62 eV) above the field-free ionization threshold.

The quantity of our interest is the width $\Gamma_{n=1}$ associated with the state originating from the ground state of the field-free hydrogen atom (from now on, we will omit the $n = 1$ label). Γ is the decay rate of the ground-state atom as a result of both the dc field ionization (tunneling) and PI by the ac field. In Fig. 1, we show the variation of Γ as a function of the dc field-strength parameter. The ac field amplitude yields $\epsilon_{ac} = 0.025$ a.u. Two important conclusions can be obtained from the exact numerical results presented in Fig. 1:

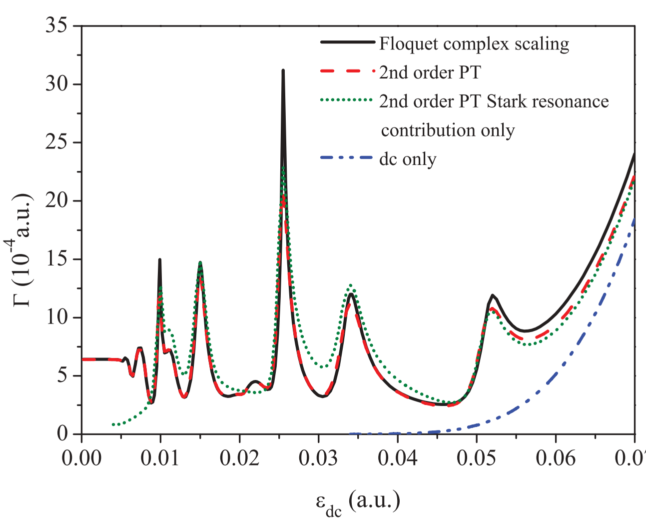

FIG. 1. (Color online) The resonance width (i.e., inverse lifetime) of the quasienergy Floquet resonance state that is issued from the 1s state of the field-free hydrogen atom by switching on the dc and ac fields. The ac field amplitude is held at $\epsilon_{ac} = 0.025$ a.u. (and $\omega = 0.633$ a.u.), whereas the dc field strength ϵ_{dc} is varied along the horizontal axis. For comparison, we show also the results obtained when only the dc field is turned on and $\epsilon_{ac} = 0$. We also show the predictions based upon the second-order perturbation theory, as discussed in the text. One observes an excellent agreement with the exact numerical results.

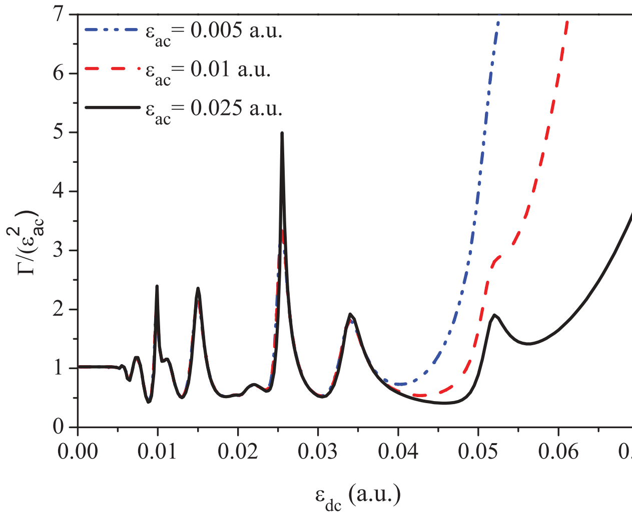

FIG. 2. (Color online) The resonance width Γ pertaining to the quasienergy Floquet resonance state shown in Fig. 1, divided by the ac field intensity. This quantity is plotted (in units of a_o^3, where a_o is the Bohr radius) as a function of the dc field strength ϵ_{dc} for three different values of ϵ_{ac}. The ac field frequency $\omega = 0.633$ a.u.

(1) By varying the strength of the weak dc field, the PI decay rate is dramatically affected, i.e., from an enhancement of the PI decay rate by a factor of 2 to 5 to a suppression of the PI process by a factor of 2 to 3.

(2) In the absence of the ac field (i.e., $\epsilon_{ac} = 0$), the lifetime of the Stark resonance state of the hydrogen atom is extremely long in comparison with the lifetime obtained when the ac field is turned on. (For $\epsilon_{ac} = 0$, the width Γ is so small that it cannot be distinguished from zero in Fig. 1 for $\epsilon_{dc} < 0.04$ a.u.)

We have repeated the same calculation for different ac field intensities, and in Fig. 2 we plot the ratio of the total rate of decay Γ versus ϵ_{ac}^2. The third conclusion obtained from the results presented in Fig. 2 is as follows:

(3) The PI rate (and therefore also the lifetime) is linearly dependent on the ac field intensity $I_0 \simeq \epsilon_{ac}^2$.

This result indicates very clearly that the corresponding complex quasienergy resonance eigenvalue can be well estimated within the framework of the second-order perturbation theory [using time in the Floquet operator as an additional coordinate (see Ref. [20])]. Here, the unperturbed Floquet Hamiltonian is given by the formula

$$\hat{\mathcal{H}}_0 = -i\hbar\partial_t - \frac{\hbar^2}{2m_e}\partial_r^2 + \frac{\hat{L}^2(\theta,\phi)}{2m_e r^2} - \frac{e^2}{r} + \epsilon_{dc} e r\cos\theta, \tag{4}$$

and the ac field is considered as a perturbation, i.e.,

$$\hat{\mathcal{H}}_1 = \epsilon_{ac} e r \cos\theta\cos(\omega t). \tag{5}$$

If so, then our PI resonance quasienergy yields

$$E_1^{dc} - \frac{i}{2}\Gamma_1^{dc} + \epsilon_{ac}^2\sum_{n\geqslant 2}\left(E_1^{(n)} - \frac{i}{2}\Gamma_1^{(n)}\right) + O\left(\epsilon_{ac}^3\right), \tag{6}$$

where the contribution of the nth Stark resonance state to the second-order perturbation expansion is given by

$$E_1^{(n)} - \frac{i}{2}\Gamma_1^{(n)} = \frac{a_n^2}{\left(E_1^{dc} + \hbar\omega - E_n^{dc}\right) - \frac{i}{2}\left(\Gamma_1^{dc} - \Gamma_n^{dc}\right)}. \tag{7}$$

The notation

$$\left\{E_n^{\mathrm{dc}} - \frac{i}{2}\Gamma_n^{\mathrm{dc}}\right\}_{n=2,\ldots}$$

stands for the complex eigenvalues of the complex-scaled Floquet operator as defined in Eqs. (1) and (2) when $\epsilon_{\mathrm{ac}} = 0$ and only the weak dc field is turned on. Correspondingly, E_n^{dc} and Γ_n^{dc} are functions of the static field-strength parameter ϵ_{dc}. Similarly, the quantities $\{\Gamma_1^{(n)}\}_{n=2,3,\ldots}$ also depend on ϵ_{dc}. The complex coefficients a_n in Eq. (7) are expressed as matrix elements

$$a_n = \left(\psi_1^{\mathrm{dc}}|er\cos\theta\cos(\omega t)e^{-i\omega t}|\psi_n^{\mathrm{dc}}\right), \tag{8}$$

where $|\psi_n^{\mathrm{dc}})$ are the Stark resonance (and possibly also continuum) eigenfunctions of the hydrogen atom in absence of the ac field, and an integration with respect to t over one optical cycle is implicitly understood. For the sake of clarity, we note explicitly in this context that an extra factor $e^{-i\omega t}$ appearing in Eq. (8) accounts for single-photon absorption processes within the framework of Floquet theory [10,20]. The results obtained by the previously described second-order perturbation theory are shown in Fig. 1 and turn out to be in remarkable agreement with the numerically exact results. The observed accuracy of second-order perturbation theory for the cases when the laser field intensity is high comes from the fact that the small parameter in our problem is not the laser intensity (which can be high), but the ratio between the laser intensity and the square of the laser frequency [see Eqs. (6) and (7)]. From the second-order perturbation analysis presented in Eqs. (6), (7), and (8), one may see that the profile of $\Gamma_1^{(n)}(\epsilon_{\mathrm{dc}})$ possesses a peak whenever the following condition is satisfied:

$$E_1^{\mathrm{dc}}\left(\epsilon_{\mathrm{dc}}^{(n)}\right) + \hbar\omega - E_n^{\mathrm{dc}}\left(\epsilon_{\mathrm{dc}}^{(n)}\right) = 0. \tag{9}$$

Here, the symbol $\epsilon_{\mathrm{dc}}^{(n)}$ stands for a specific dc field strength at which the requirement of Eq. (9) is fulfilled for a given Stark resonance n. At the dc field equal to $\epsilon_{\mathrm{dc}}^{(n)}$, one may argue that a maximal enhancement of the PI rate should be observed, given by a single dominant term of Eq. (6).

Our numerical results presented in Fig. 3 confirm that the previously discussed theoretical arguments indeed give an adequate physical picture of the PI enhancement phenomenon. Namely, PI maxima do appear precisely at those dc field strengths predicted by Eq. (9). In the upper panel of Fig. 3, the condition of Eq. (9) corresponds to the points where the $n = 1$ level shifted by the photon energy $\hbar\omega$ crosses the Stark-Rydberg resonance levels emerging from lower-lying excited bound states of the field-free hydrogen atom. (For the sake of clarity, we point out in passing that only those Stark resonances that are driven by the dc field to the one-photon excitation region are plotted in Fig. 3. The other Stark resonances, although present in our computations, are not shown.) The emerging conclusion is clear: An enhancement of the PI decay rate is obtained whenever the dc field-strength parameter is such that the ground state of hydrogen (hardly affected by the dc field) is excited to one of the excited Rydberg-Stark resonances of the hydrogen atom, and the second-order perturbational expansion of the PI decay rate is dominated by one term only. The latter situation is demonstrated in the lower panel of Fig. 3, where the contributions of the individual Stark resonances $\{\Gamma_1^{(n)}\}_{n=2,\ldots}$, as defined in Eq. (7), are also shown. Evidently, they contribute dominantly to the appropriate peaks.

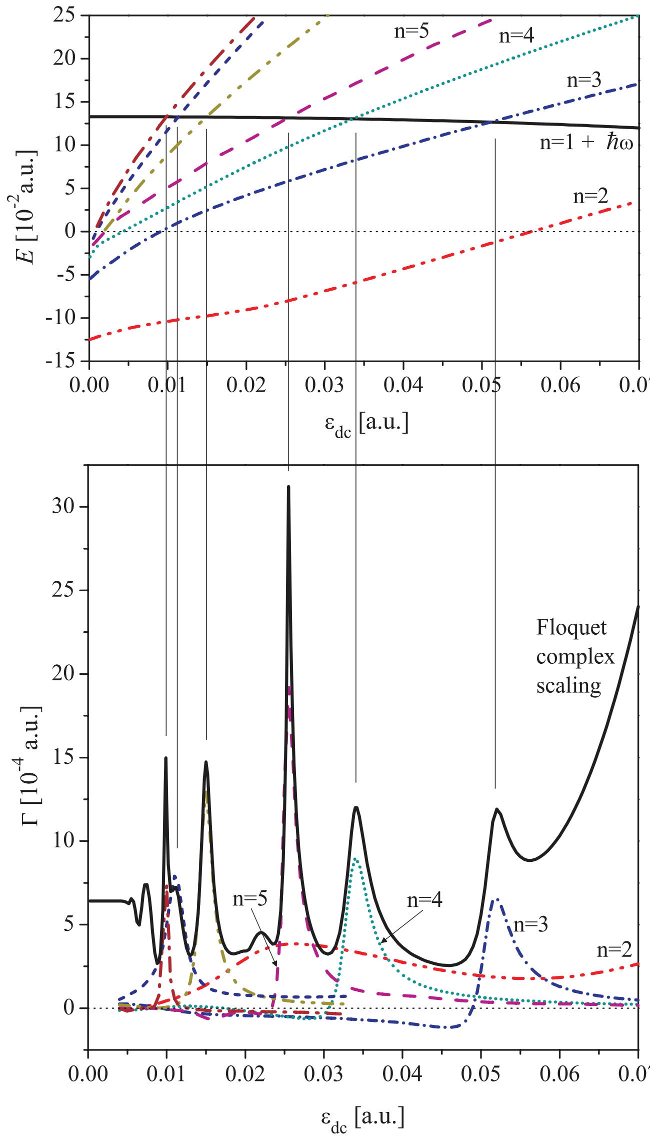

FIG. 3. (Color online) Upper panel: The Stark resonance energies of the hydrogen atom (no ac field) plotted as functions of the dc field-strength parameter ϵ_{dc}. The position of the Stark resonance that is associated with the ground state of the hydrogen atom ($n = 1$) was shifted by one photon energy (by $\omega = 0.633$ a.u.). The dotted horizontal line at zero energy is the ionization threshold of the field-free hydrogen. Lower panel: The numerically exact values of the decay rate Γ of the quasienergy Floquet resonance state that is associated with the ground field-free state of the hydrogen atom, plotted as a function of the dc field-strength parameter ϵ_{dc}, when $\epsilon_{\mathrm{ac}} = 0.025$ a.u. (solid line). Broken and dotted lines depict here the individual contributions $\Gamma_1^{(n)}$ to the PI rate as a result of different excited Stark resonance states [see Eq. (7)].

Let us discuss now the last part of our perturbational analysis, which is focused on the suppression of PI by the weak dc field. As seen in Fig. 3, the situation of $\Gamma(\epsilon_{\mathrm{ac}},\epsilon_{\mathrm{dc}}^{(k)}) \ll \Gamma(\epsilon_{\mathrm{ac}},\epsilon_{\mathrm{dc}} = 0)$ occurs when the dc field-strength parameter ϵ_{dc} is chosen roughly in-between two adjacent peaks. Since the peaks in the PI spectra shown in Figs. 1, 2, and 3 are consequences of the photoexcitation by the ac field from the ground state to the excited Rydberg-Stark resonance states, it seems very reasonable to assume that the suppression of the PI rate of decay arises because of an interference between two adjacent Stark resonances, which are coupled by the ac field. This conjecture is fully confirmed by the lower panel in Fig. 3. Namely, the individual contributions $\Gamma_1^{(n)}$ possess negative values far from the peaks, and the sum of the adjacent terms $\Gamma_1^{(n-1)}$ and $\Gamma_1^{(n)}$ gets values that are smaller than the PI rate of decay, which is obtained at $\epsilon_{\rm dc} = 0$.

Finally, let us briefly summarize the contents of this paper. We can conclude that, even for the high-intensity regime of the ac field, the second-order perturbation theory explains quantitatively how the PI decay rate is enhanced or suppressed by a weak dc field. The ac field can be regarded as a perturbation whenever its frequency is sufficiently high. An enhancement of the PI rate of decay results from the photoexcitation of the ground state of hydrogen to one of the excited Stark resonance states. On the other hand, a destructive interference takes place between two adjacent Stark resonances for certain values of the dc field-strength parameter, and the PI rate of decay is suppressed. It is expected that the previously described physical picture is also applicable to other atoms.

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[1] L. Gallmann, Nature Phys. **6**, 406 (2010).
[2] L. Young *et al.*, Nature (London) **466**, 56 (2010).
[3] U. Eichmann, T. Nubbemeyer, H. Rottke, and W. Sandner, Nature (London) **461**, 1261 (2009).
[4] M. P. de Boer and H. G. Muller, Phys. Rev. Lett. **68**, 2747 (1992).
[5] N. Moiseyev and F. Weinhold, Phys. Rev. Lett. **78**, 2100 (1997).
[6] A. L'Huillier, K. J. Schafer, and K. C. Kulander, J. Phys. B **24**, 3315 (1991).
[7] K. Burnett, V. C. Reed, and P. L. Knight, J. Phys. B **26**, 561 (1993).
[8] I. A. Ivanov and A. S. Kheifets, Eur. Phys. J. D **38**, 471 (2006).
[9] S. Cocke and L. E. Reichl, Phys. Rev. A **52**, 4515 (1995).
[10] I. Gilary *et al.*, Phys. Rev. Lett. **101**, 163002 (2008).
[11] A. M. Popov, O. V. Tikhonova, and E. A. Volkova, J. Phys. B **36**, R125 (2003).
[12] M. V. Fedorov, *Progress in Ultrafast Intense Laser Science* (Springer, Berlin, 2006), Vol. I.
[13] M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990).
[14] M. V. Fedorov and N. P. Poluektov, Phys. Rev. A **69**, 033404 (2004).
[15] N. Moiseyev, Phys. Rep. **302**, 212 (1998).
[16] S. I. Chu, Adv. At. Mol. Phys. **21**, 197 (1985).
[17] E. Balslev and J. M. Combes, Commun. Math. Phys. **22**, 280 (1971).
[18] B. Simon, Commun. Math. Phys. **27**, 1 (1972); Int. J. Quantum Chem. **14**, 529 (1978).
[19] Because of an axial symmetry (the laser field is polarized along the static field direction), M is a good quantum number. Since the ground state is $M = 0$, we use only $M = 0$ states in our computation.
[20] H. Sambe, Phys. Rev. A **7**, 2203 (1973); J. Howland, Math. Ann. **207**, 315 (1974); U. Peskin and N. Moiseyev, J. Chem. Phys. **99**, 4590 (1993).