Suppression of Photoionization by a Static Field

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(Received 19 December 2007; published 17 October 2008)

The dc field Stark effect is studied theoretically for atoms in high intensity laser fields. We prove that the first-order perturbation corrections for the energy and photoionization rate vanish when the dc field strength serves as a perturbational strength parameter. Our calculations show that by applying a dc field in the same direction as the polarization direction of the ac field, the photoinduced ionization rate is almost entirely suppressed. This suppression is attributed to changes in the phase shift of the continuum atomic wave functions which can be controlled by the dc field.

DOI: 10.1103/PhysRevLett.101.163002 PACS numbers: 32.60.+i

One of the crucial factors for improving the efficiency of many light induced processes is to lower the photoionization (PI) rate. Therefore, the suppression of photoionization has been the focus of numerous theoretical and experimental investigations (see Refs. [1,2] and references therein). Among the various stabilization mechanisms, let us mention the Kramers-Henneberger stabilization (KHS) [3] and interference stabilization (IS) [4]. 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 IS, on the other hand, requires an interaction of several Rydberg states.

Former studies of stabilization mechanisms were mainly concerned with changing the natural behavior by which the PI rate is varied with the laser frequency. A Cooper minimum [5,6] is a dip, sometimes reaching zero, in the PI cross section of an atom appearing at a certain photon energy due to special combination of symmetries of the ground state and the continuum wave function at the corresponding energy above threshold. As we will show here the mechanism we propose for suppression of PI is different and very robust. We will show that it holds for 1D and 3D potentials, short range potentials, as well as for a long range Coulombic potential.

Using a semiclassical approach and the quantum mechanical second-order perturbation theory, Cocke and Reichl [7] have shown that, by adding a static field to a laser driven δ-function potential, one can stabilize the PI rate for "a selected range of frequencies of the incident laser field." In the present Letter we extend this idea beyond the framework of perturbation theory and propose a mechanism that allows the suppression of the PI rate in a *broad and continuous* range of field intensities and frequencies by applying an additional dc field. This somewhat counterintuitive mechanism is the result of the quantum mechanical nature of the ionization process, and, therefore, is very interesting from a theoretical viewpoint. The possibility of controlling the PI rate by an additional external parameter is also appealing from an experimental point of view. Experimentally the static field can be emulated by adding a second low frequency beam to the main laser beam.

For the sake of simplicity we first discuss the suppression of PI by a static field for a 1D potential which has been used extensively in the literature for the study of the interaction of Xe atoms with a laser field [8]. Then we will introduce the applicability of our approach to an effective 3D potential which describes the same physical system of Xe in the presence of both ac and dc fields.

The corresponding Hamiltonian, expressed in atomic units ($\hbar = e = m = 1$), takes the form

$$\hat{H}(x, t) = -\partial_x^2/2 + V(x) - x[\varepsilon_{\rm ac}\cos(\omega_L t) + \varepsilon_{\rm dc}]. \quad (1)$$

Here, $V(x) = -0.63\exp(-0.1424x^2)$ stands for an effective potential function where the parameters were fit such that the two lowest energy levels $E_0 = -0.4451$ a.u. and $E_1 = -0.1400$ a.u. will reproduce the ionization energies of the ground and first excited states of xenon. Thus these states mimic the energies of the full many-electron levels of the atom with respect to the first ionization threshold.

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Unless stated otherwise the laser frequency is $\omega_L = 0.5$ a.u..

Before we delve into the numerical results for the model presented above, we wish to give some physical intuition as to why one can control the PI rate by adding a dc field. In the absence of the dc field, the resonances are the result of the ac field induced interaction between the bound states of the field-free potential and the continua of the field-free potentials shifted down by $n\hbar\omega$. This is the so-called dressed (Floquet) picture which will be discussed below in more detail. We restrict the dc field strength so that the ground state wave function of the field-free potential is hardly affected by ε_{dc}. Therefore, by adding the dc field we only change the structure of the continua of the field-free potential which will then be coupled to the ground state via the ac field. Such a change in the continua can lead to enhancement or suppression of the PI rate. We shall come back to this point later in the letter.

Since the Hamiltonian (1) is time periodic with the period $T = 2\pi/\omega_L$, the ionization rates we seek can be calculated by employing the complex scaled Floquet theory [9,10]. Using complex scaling the photoinduced dynamics is associated with a single quasienergy (Floquet) resonance eigenstate rather than with a wave packet as in the standard (Hermitian) formalism of quantum mechanics. Within the theoretical framework of Floquet theory, the time variable t is treated as an additional dynamical coordinate and the corresponding Floquet Hamiltonian is given by

$$\hat{\mathcal{H}}(x, t) = \hat{H}(x, t) - \imath\hbar\partial_t. \tag{2}$$

In order to describe efficiently the ionization phenomena and to calculate the decay rates of metastable (resonance) photoionized states the coordinate x is rotated into the complex plane ($x \to xe^{i\theta}$). The Floquet states are given by $\Psi_\alpha^\theta(x, t) = \exp(-i\mathcal{E}_\alpha^\theta t/\hbar)\Phi_\alpha^\theta(x, t)$, where $\Phi_\alpha^\theta(x, t) = \Phi_\alpha^\theta(x, t + T) = \sum_{n=-\infty}^{n=+\infty} \varphi_{n,\alpha}^\theta(x)\exp(+in\omega_L t)$ are the eigenstates of the Floquet Hamiltonian $\hat{\mathcal{H}}$ in (2) with corresponding eigenvalues $\mathcal{E}_\alpha^\theta$.

The Floquet states given above can be divided into two classes. One type of solutions are the photoinduced resonances, for which the quasienergy $\mathcal{E}_\alpha^\theta$ is θ independent, and $\Gamma_\alpha = -2Im[\mathcal{E}_\alpha^\theta]$ is the corresponding PI decay rate. In our notation, the ($\alpha = 0$) resonance state corresponds to the field-free ground state of $V(x)$ in the limit when both the ac and the dc fields are adiabatically switched off. The second type of solutions are the rotated continuum states, whose quasienergies are $\mathcal{E}_\alpha^\theta = |\mathcal{E}_\alpha|\exp(-i2\theta)$. Since we are interested in the first type of solutions which are not dependent on the scaling angle the notation θ will be dropped from here on.

Numerically exact Floquet calculations for the model 1D potential given above have been performed by using particle-in-a-box basis functions for x, and by propagating using the t, t' method [11]. Figure 1 displays the obtained PI rate $\Gamma_{\alpha=0}$ scaled by ε_{ac}^2 and plotted vs the dc field strength ε_{dc}. We show the results for three different values of ε_{ac}. As can be seen if Fig. 1 the PI is almost completely suppressed without being very sensitive to small variations of the dc field strength parameter, for $\varepsilon_{dc} \approx 0.01$ a.u. without dependence on the laser field intensity, which varies dramatically. This is the most important result of the present Letter, which as will be shown later holds also for 3D potentials. The notably high suppression of the PI induced by the laser can be obtained for any value of $\omega_L > 0.45$ a.u. (where single photon processes are dominant) when the static field reaches a frequency dependent critical value.

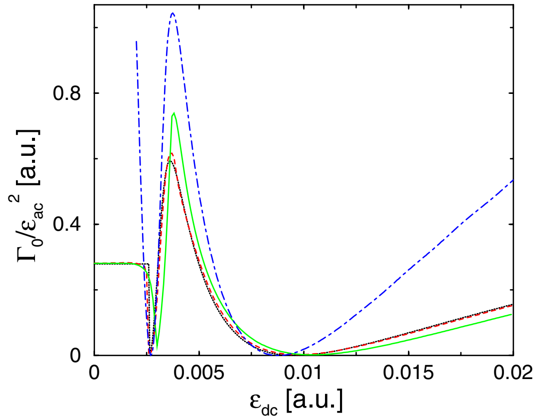

FIG. 1 (color online). The ratio $\Gamma_0/\varepsilon_{ac}^2$ plotted vs the dc field strength ε_{dc}. Γ_0 is the photoionization rate as obtained from our numerically exact Floquet calculations. The dotted line is for $\varepsilon_{ac} = 0.005$ a.u., the dashed line for $\varepsilon_{ac} = 0.035$ a.u., while the solid line is for $\varepsilon_{ac} = 0.075$ a.u.. Note that the ionization rate is totally suppressed regardless of the value of ε_{ac}. The dot-dashed line represents the results of the golden rule formula in (3).

Before exploring the origin of this result, we would like to analyze the curve presented in Fig. 1 in the limit of $\varepsilon_{dc} \to 0$ where no suppression of the PI rate occurs. The effect of the dc field on $\Gamma_{\alpha=0}$ at the limit of $\varepsilon_{dc} \to 0$ can be accounted for by a perturbational analysis within the Floquet treatment of the system with ε_{dc} as a small parameter. Such an approach can be regarded as an extension of the usual perturbative treatment of the dc field Stark effect for atoms in the absence of laser light [12]. The zero-order states are the Floquet solutions denoted by $\Psi_\alpha^0(x, t) = \exp(-i\mathcal{E}_\alpha^0 t/\hbar)\Phi_\alpha^0(x, t)$ which are obtained in the absence of the dc field, i.e., $\varepsilon_{dc} = 0$, where $\mathcal{E}_\alpha^0 = E_\alpha^0 - i\Gamma_\alpha^0/2$. Therefore, the zeroth-order energy and PI rate of the Floquet resonance state which is associated with the ground state of the field-free atom are given by E_0^0 and Γ_0^0, respectively. One should be aware of the fact that due to the presence of the laser field $\Gamma_0^0 \neq 0$.

The first-order correction to $\mathcal{E}_0 = E_0 - i\Gamma_0$ is given by $\mathcal{E}_0^1 = \langle\langle\Phi_0^0|\varepsilon_{dc}x|\Phi_0^0\rangle\rangle$ where the notation $\langle\langle\cdot\cdot\rangle\rangle$ implies integration over both space and over one cycle period of the laser field. This correction vanishes due to the symmetry of the Fourier components of the Floquet solutions and of x. To demonstrate this last point one must observe that the

Floquet Hamiltonian in (2) is invariant under the following transformation: $x \to -x, t \to t + T/2$. This means that the eigenstates of $\hat{\mathcal{H}}$, i.e., Φ_α^0 must also possess the same symmetry. Therefore $\Phi_\alpha^0(-x, t + T/2) = \pm\Phi_\alpha^0(x, t)$. In terms of the Fourier components $\varphi_{n,\alpha}$ of Φ_α^0 this implies that $\pm\varphi_{n,\alpha}(x) = (-1)^n \varphi_{n,\alpha}(-x)$. The perturbation in this case is the dipole x which is time independent; therefore, the integration over time reduces the first-order correction $\mathcal{E}_0$ to $\mathcal{E}_0^1 = \varepsilon_{\mathrm{dc}}\sum_n \langle\varphi_{n,0}|x|\varphi_{n,0}\rangle$. Since the Fourier components $\varphi_{n,0}$ have distinct parity and x is an odd function the first-order correction to $\mathcal{E}_0$ vanishes. This result demonstrates that the dc field Stark effect is quadratic in $\varepsilon_{\mathrm{dc}}$ even in the case of atoms in laser fields where all bound states become metastable. To the best of our knowledge this extension of the Stark effect to the rate of decay of metastable states is new.

Let us now return to the most dramatic result of this Letter. The prominent suppression of the PI rate demonstrated in Fig. 1 as well as its robustness with respect to the laser field intensity is very surprising indeed. As will be shown, this phenomenon has its origin in the quantum mechanical nature of the ionization process. As mentioned above, within the dressed picture the light induced dynamics of an atom can be described as a coupled channel problem. The diagonal channel potentials are $V(x) - \varepsilon_{\mathrm{dc}}x + m\hbar\omega_L$, with the so-called Brillouin zone index m ranging from $-\infty$ to $+\infty$. A given channel m is coupled with neighboring channels $m \pm 1$ through a coupling term $\varepsilon_{\mathrm{ac}}x/2$. As already discussed, the ionization occurs due to the coupling between the bound states of the diagonal potential with the continua of the other channels. Although in general one must take into account the coupling with all channels, usually the coupling is strongest between the neighboring channels. If we account only for the decay of the ground state of the potential into the continuum of the closest underlying channel, the rate of decay can be calculated using the well-known golden rule formula [13], which represents in fact an outcome of the second-order perturbation theory in $\varepsilon_{\mathrm{ac}}$. The golden rule PI rate then reads

$$\Gamma(\varepsilon_{\mathrm{dc}}) = \left(\frac{\varepsilon_{\mathrm{ac}}}{2}\right)^2 2\pi|\langle\psi_0(\varepsilon_{\mathrm{dc}})|x|\psi_{E_0+\hbar\omega_L}(\varepsilon_{\mathrm{dc}})\rangle|^2, \qquad (3)$$

where $\psi_0(\varepsilon_{\mathrm{dc}})$ is the ground state wave function of the diagonal potential and $\psi_{E_0+\hbar\omega_L}(\varepsilon_{\mathrm{dc}})$ is the continuum state of the lower channel at the energy of the ground state of the upper channel. An immediate observation of the expression in (3) reveals that the PI rate is proportional to $\varepsilon_{\mathrm{ac}}^2$ just as we saw in the numerically exact results presented in Fig. 1. Thus, the characteristic curve of the normalized PI rate shown in Fig. 1 clearly shows that the main contribution to the ionization rate comes here from the single photon decay of the ac field-free bound state into the corresponding continuum state of the lower channel. The dependence of the golden rule decay rate in (3) on $\varepsilon_{\mathrm{dc}}$ is shown in Fig. 1 by the dot-dashed line and is in excellent qualitative agreement with the exact numerical results which take into account all the coupled channels. Since the same behavior for various ac field strengths is readily explained from the golden rule rate, the mechanism of enhancement and suppression of the PI for specific dc field strengths seems to be associated with the properties of the continuum functions in the region where the bound state of the atom is localized. We discuss this point below in more detail.

If the binding energy of the ground state is large enough, and the nearest excited state is far enough, then the addition of an external dc field will hardly affect the ground state wave function. This is not the case for the continuum of the field-free potential which changes due to the additional static field. Therefore, the origin of suppression of the PI rate can now be traced using (3). We have already established that the only term in (3) that is modified by the static field is the continuum wave function. Its phase shift changes as the strength of the applied static field increases and the corresponding matrix element of (3) eventually changes sign. Therefore, at least within the golden rule expression, complete suppression of PI is achievable. Note that this has been observed before in special cases [7] when the dipole matrix elements are known analytically.

The addition of the static field to the field-free Hamiltonian breaks the symmetry. However, since the ground state wave function is hardly affected by this change it more or less maintains its symmetry properties. Therefore, in order to suppress the PI rate the appropriate continuum state should be as close as possible to an even function in the local range occupied by the ground state wave function. Only then the matrix element vanishes (the dipole moment is an odd function in the spatial coordinate). Enhancement of the PI rate is achieved in the opposite case where the continuum wave function attains a local odd parity in the region of the potential occupied by the ground state. On the basis of this explanation it may happen that multiple local minima in the photoionization decay rate as function of strength of the static field might be observed. As we will show below this is indeed so in some cases. The validity of this analysis has been checked by examining the continuum wave function in (3) at the point where suppression is attained in Fig. 1. Indeed, at this static field strength the ground state $|\psi_0\rangle$ had an almost perfect even symmetry and was localized in the region where the continuum function $|\psi_{E_0+\hbar\omega_L}\rangle$ has a local even parity property.

Is this mechanism of suppressing PI by dc field applicable to 3D potentials? As we will show below the answer to this question is positive. The reason is as follows. The Coulombic field-free atomic potential $V_{\mathrm{atom}}(r)$ has a spherical symmetry. The addition of an electromagnetic field breaks this symmetry. However, when the dc field is introduced along the same axis where the laser field is linearly polarized the breaking of the symmetry will occur only in that direction. Thus the general behavior should be

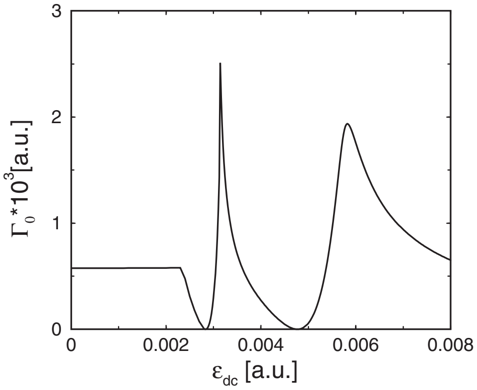

FIG. 2. The dependence of the PI decay rate of a model 3D xenon atom in a combination of a cw laser field and a static field on the dc field strength. An electron in the field-free atom in this model is assumed to be moving under the influence of an effective radial potential of the form $V(r) = -V_0/\cosh^2(ar)$, where $V_0 = 1.074$ a.u. and $a = 2.814$ a.u.. The laser field strength is $\epsilon_{ac} = 0.035$ a.u. while the laser frequency is $\omega_L = 0.5$ a.u. throughout.

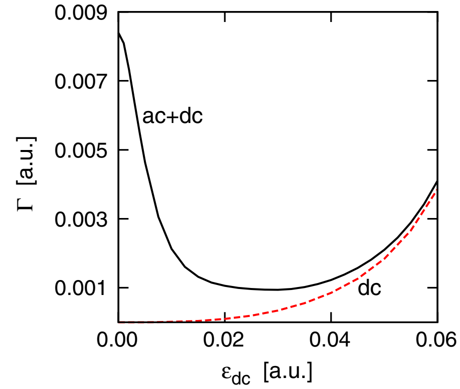

FIG. 3 (color online). The dependence of the PI decay rate of hydrogen atom in a combination of a cw laser field and a static field on the dc field strength. The laser field strength is $\epsilon_{ac} = 0.025$ a.u. and the laser frequency is $\omega_L = 0.6330$ a.u. throughout. The ionization decay rate due to the dc field alone is added for comparison in the dashed line.

similar to the one-dimensional case with additional features due to the motion in the other directions. In this sense it is significant that the one-dimensional model chosen earlier to describe the suppression of the PI is symmetrical since when the spherical symmetry of the atom is broken it is along the laser polarization axis where initially the potential was symmetrical. This makes the analogy between the cases much more sensible rather than a nonsymmetrical 1D potential.

In order to support these arguments, we have carried out numerical calculations of the photoinduced dynamics for the case of the 3D Rosen-Morse potential, $V(r) = -V_0/\cosh^2(ar)$. The potential parameters were fit to mimic the energies of the ground (s-type) state and the first excited (p-type) state of Xe just as in the one-dimensional case. In Fig. 2 we show the photoinduced ionization rate as obtained from our 3D calculations. A comparison with the previously discussed rate obtained using the 1D approach shows an overall qualitative agreement. In both cases there is an abrupt suppression of the photoinduced ionization rate at $\epsilon_{dc} \sim 0.0028$ a.u. and another suppression that is gradually obtained at a stronger dc field which is the focus of the present study. The differences result from the bound-bound transitions between the field-free 3D states which are induced by the dc field and by the fact that in 3D the photoionization resonance state has several open channels for decay. Yet, the explanation given above based on the change of the *local* symmetry property of the continuum wave function as ε_{dc} is varied holds for the 3D case just as well as for the one-dimensional case.

The last question we should address is how robust is the suppression mechanism presented here? Is it restricted to short range potentials that we have studied here or can it be observed also for long range Coulombic potential? A long time ago Chu and Cooper [14] calculated the PI rate of decay of a hydrogen atom in an intense laser field. We reproduced the results of Ref. [14] using for the calculation a d-aug-CC-$pV6Z$ basis set and extended their approach to include the simultaneous interaction of a hydrogen atom with both ac and dc fields. The results presented in Fig. 3 show that even for real *ab initio* calculations the addition of the static field induces suppression just as obtained before for the 1D and 3D model Hamiltonians of xenon.

We conclude by restating that the mechanism of PI suppression by a static field due to the continuum phase shift is a robust phenomenon which is obtained for short range 1D and 3D potentials, as well as for a long range Coulombic potential as has been shown here by our *ab initio* calculations for the hydrogen atom.

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[1] A. M. Popov, O. V. Tikhonova, and E. A. Volkova, J. Phys. B **36**, R125 (2003).
[2] M. V. Fedorov, *Progress in Ultrafast Intense Laser Science* (Springer, Berlin, Heidelberg, 2006), Vol. I.
[3] M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990).
[4] M. V. Fedorov and N. P. Poluektov, Phys. Rev. A, **69**, 033404 (2004).
[5] J. W. Cooper, Phys. Rev. **128**, 681 (1962).
[6] U. Fano and J. W. Cooper, Rev. Mod. Phys. **40**, 441 (1968).
[7] S. Cocke and L. E. Reichl, Phys. Rev. A **52**, 4515 (1995).
[8] J. N. Bardsley and M. J. Comella, Phys. Rev. A **39**, 2252 (1989).
[9] W. P. Reinhardt, Annu. Rev. Phys. Chem. **33**, 223 (1982).
[10] N. Moiseyev, Phys. Rep. **302**, 212 (1998).
[11] U. Peskin and N. Moiseyev, J. Chem. Phys. **99**, 4590 (1993).
[12] J. Stark, Ann. Phys. (Leipzig) **348**, 965 (1914).
[13] W. H. Miller, Chem. Phys. Lett. **4**, 627 (1970).
[14] S.-I. Chu and J. Cooper, Phys. Rev. A **32**, 2769 (1985).