Conditions for the applicability of the Kramers-Henneberger approximation for atoms in high-frequency strong laser fields

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Using well-known scaling techniques we present that for a given laser intensity the photoionization decay rate Γ of the core electron in the $1s$ orbital of an atom is varied with the laser frequency ω such that $\Gamma \mathcal{Z}^4$ vs $\omega/\mathcal{Z}^2$ has a universal behavior that does not depend on the nuclear charge $\mathcal{Z}$. One can conclude that the laser frequency required to stabilize the $1s$ core electron is larger by the factor $\mathcal{Z}^2$ than the laser frequency required to stabilize the electron in the most diffused orbital. This condition ensures the applicability of the Kramers-Henneberger approximation (KHA) for all electrons including the core electrons. However, simple analytical arguments, which are confirmed by our numerical results, show that when the photoionization of the core electron is due to the absorption of one photon only, then the KHA is applicable for a lower frequency that is linearly proportional to $\mathcal{Z}$ and not to the square of atomic number as expected. This condition is a lower bound for the laser frequency which is needed to stabilize a many-electron atom in the presence of high-frequency strong laser fields.

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Presently high-intensity laser pulses with a dominant frequency which is higher than the atomic frequency of the valence electrons in atoms are feasible and enable applications in different areas of physics. One may expect that strong laser fields always lead to a fast photoionization process. As was shown recently by Eichmann *et al.* [1,2], most of the atoms which were accelerated by intense femtosecond laser pulses remained stable during the pulse. The electronic structure of these stable "laser-dressed" atoms can be visualized by spectroscopy of photoelectrons [3]. The question of the applicability of the Kramers-Henneberger approximation (KHA) [4,5] which provides the picture of stable "laser-dressed" atoms in the theory of strong-field ionization has been a subject of research for a long time [6–17]. In the most recent publications on chemistry and physics in strong high-frequency laser fields, it has been presented that by applying the KHA for all electrons including the core electrons the interesting phenomena can be observed, e.g., strong chemical bonds in He_2 and HeS molecules [18,19], a strong linear Stark effect for a sulfur atom rather than the usual quadratic one [20].

The KHA is based on the acceleration gauge representation of the problem. However, when the length gauge is used instead of the acceleration gauge the core electrons are often assumed not to be affected by the external field. It means that only the valence electrons "feel" the oscillating laser field. Here we address the question under what condition one should apply the KHA not only for the electrons in the valence shell, but also for the core electrons. No doubt, the answer to this question is important for new developed areas of chemistry and physics in high-frequency strong laser fields. In particular, due to the enormous investments in free-electron laser equipment, it is possible to carry out experiments in a high-frequency and large intensity regime which could not be done before.

In this paper, we take a single-electron approach: Each one of the two core electrons in the $1s$ orbital is described by a one-electron Hamiltonian, where the nuclear charge is $\mathcal{Z}$.

Here we neglect the small screening effect of two electrons occupying the same orbital. For a given laser intensity we look for the minimal laser frequency for which the core electrons are stabilized and the Kramers-Henneberger approximation holds. This value of the frequency provides a lower bound to the actual laser frequency that stabilizes the core electrons in the many-electron neutral atom. Based on Gauss's electrostatic law, we know that the effective nuclear charge for the electron in the most diffused orbital, which is outside of a sphere containing all the other electrons, is 1, i.e., $\mathcal{Z}_{\text{eff}} = 1$. According to the Slater's rules for the valence electron of an atom $\mathcal{Z}_{\text{eff}} > 1$, but it is much smaller than $\mathcal{Z}$, i.e., $\mathcal{Z}_{\text{eff}} \ll \mathcal{Z}$ [21]. This is the base for our claim that a comparison between the photoionization decay rate of a one-electron ion with the nuclear charge $\mathcal{Z} > 1$ and the photoionization decay rate of a hydrogen atom ($\mathcal{Z} = 1$) gives us the ratio between the decay rate of the $1s$ core electron and the decay rate of the valence electron that populates the most diffused atomic orbital of a many-electron atom.

We use the well-known $\mathcal{Z}$-scaling technique. For field-free many-electron atoms, it has been applied by Hylleraas to develop the $1/\mathcal{Z}$ perturbation theory [22]. For example, Bielińska-Wąż *et al.* have used such a technique to investigate spectra of hydrogenic systems embedded in a Debye plasma environment [23]. The $\mathcal{Z}$ scaling has also been used many years ago by Madsen and Lambropoulos in the their study of the strong-field ionization of hydrogenlike atoms [24]. They have found some general scaling relations perturbative or directly from the time-dependent Schrödinger equation. Our approach is slightly different. We do not scale time by the nuclear charge $\mathcal{Z}$ as in Ref. [24], but define dimensionless time variable, $\tau = \omega t$. This is a crucial point in our derivation presented below. By using the $\mathcal{Z}$ scaling we first show that the photoionization decay rate Γ of the $1s$ core electron of an atom with a nuclear charge $\mathcal{Z}$ is varied with the frequency ω of a strong linearly polarized laser field such that $\Gamma \mathcal{Z}^4$ vs $\omega/\mathcal{Z}^2$ has a universal functional behavior which is $\mathcal{Z}$ independent.

The most important result of our derivation is the new scaling law: For a given laser intensity the ratio between the decay rate of the electron in the most diffused orbital and the decay rate of the electron in the $1s$ orbital of an atom is equal to $\mathcal{Z}^4$, while the laser frequency which is required to stabilize the core electron (i.e., the KHA is applicable) is larger by the factor $\mathcal{Z}^2$ than the frequency necessary to stabilize the valence electron. However, our numerical *ab initio* calculations and simple analytical analysis show that this condition for stability is too strong. Actually, it is sufficient to stabilize the $1s$ core electron by the laser frequency which is larger by the factor $\mathcal{Z}$ than the frequency required to stabilize the electron in the most diffused orbital where $\mathcal{Z}_{\text{eff}} \simeq 1$. This is another important conclusion of our work.

Let us consider an atom in an external ac field within the frameworks of the length gauge and the acceleration gauge representations. In the dipole approximation the time-dependent Schrödinger equation (TDSE) for a single-electron system in a linearly polarized strong laser field in the length gauge representation has the form

$$\left[-\frac{\hbar^2}{2m}\nabla_{x,y,z}^2 - \frac{\mathcal{Z}e^2}{\sqrt{x^2+y^2+z^2}} + e\varepsilon_0 z f(t)\cos(\omega t) \right]$$
$$\times |\psi(t)\rangle = i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle, \tag{1}$$

where e and $\mathcal{Z}e$ are, respectively, the charges of the electron and the nucleus. The shape of the laser envelope is given by $f(t)$. The laser intensity is defined as $I_0 \propto \varepsilon_0^2$, where ε_0 is the field amplitude. By carrying out the transformation $\{x,y,z\} \to \mathcal{Z}\{x,y,z\}$, one obtains the following TDSE:

$$\left[-\frac{\hbar^2}{2m}\nabla_{x,y,z}^2 - \frac{e^2}{\sqrt{x^2+y^2+z^2}} + e\frac{\varepsilon_0}{\mathcal{Z}^3} z f(t)\cos(\omega t) \right]$$
$$\times |\psi(t)\rangle = i\frac{\hbar}{\mathcal{Z}^2}\frac{\partial}{\partial t}|\psi(t)\rangle. \tag{2}$$

From Eq. (2), it is clear that as $\mathcal{Z}$ gets larger values the first-order perturbation theory becomes a better approximation when the zero-order Hamiltonian is the field-free Hamiltonian of a hydrogen atom. Let us now define dimensionless time unit

$$\tau = \omega t, \quad 0 \leqslant \tau \leqslant 2\pi, \tag{3}$$

such that Eq. (2) is transformed into

$$\left[-\frac{\hbar^2}{2m}\nabla_{x,y,z}^2 - \frac{e^2}{\sqrt{x^2+y^2+z^2}} + e\varepsilon_0^{\text{scaled}} z\, g(\tau)\cos(\tau) \right]$$
$$\times |\phi(\tau)\rangle = i\hbar\omega_{\text{scaled}}\frac{\partial}{\partial\tau}|\phi(\tau)\rangle, \tag{4}$$

where

$$|\phi(\tau)\rangle = |\psi(t=\tau/\omega)\rangle, \tag{5}$$

$$g(\tau) = f(t=\tau/\omega) \tag{6}$$

and

$$\omega_{\text{scaled}} = \frac{\omega}{\mathcal{Z}^2}, \tag{7}$$

$$\varepsilon_0^{\text{scaled}} = \frac{\varepsilon_0}{\mathcal{Z}^3}. \tag{8}$$

By following the same scaling transformation procedure in the acceleration gauge (ag) representation [4,5,25–29], one obtains

$$\left[-\frac{\hbar^2}{2m}\nabla_{x,y,z}^2 - \frac{e^2}{\sqrt{x^2+y^2+\left[z - e\varepsilon_0^{\text{scaled}}m^{-1}\omega_{\text{scaled}}^{-2}\int_0^\tau d\tau'\int_0^{\tau'} d\tau'' g(\tau'')\cos(\tau'')\right]^2}} \right]|\phi_{\text{ag}}(\tau)\rangle = i\hbar\omega_{\text{scaled}}\frac{\partial}{\partial\tau}|\phi_{\text{ag}}(\tau)\rangle. \tag{9}$$

Within the Kramers-Henneberger approximation the laser-dressed atom is well described by the effective time-independent Hamiltonian which includes the time-averaged potential. The electron in such potential oscillates because of its strong interaction with the electromagnetic field. The Kramers-Henneberger (KH) Hamiltonian [4,5] after our transformation has the form

$$\hat{H}_{\text{KH}} = -\frac{\hbar^2}{2m}\nabla_{x,y,z}^2 - \frac{1}{2\pi}\int_0^{2\pi} d\tau \frac{e^2}{\sqrt{x^2+y^2+\left[z - e\varepsilon_0^{\text{scaled}}m^{-1}\omega_{\text{scaled}}^{-2}\int_0^\tau d\tau'\int_0^{\tau'} d\tau'' g(\tau'')\cos(\tau'')\right]^2}}. \tag{10}$$

For a laser pulse whose duration is sufficiently long the laser envelope can be considered as unity, $g(\tau) = 1$ [and $f(t) = 1$] [30]. Therefore, the above KH Hamiltonian is reduced to

$$\hat{H}_{\text{KH}} = -\frac{\hbar^2}{2m}\nabla_{x,y,z}^2 - \frac{1}{2\pi}\int_0^{2\pi} d\tau$$
$$\times \frac{e^2}{\sqrt{x^2+y^2+\left[z+\alpha_0^{\text{scaled}}\cos(\tau)\right]^2}}, \tag{11}$$

where α_0^{scaled} is the quiver length defined as

$$\alpha_0^{\text{scaled}} = \frac{e\varepsilon_0^{\text{scaled}}}{m\omega_{\text{scaled}}^2}. \tag{12}$$

The applicability of the Kramers-Henneberger approximation, where the dressed Hamiltonian is time independent, depends on the laser parameters for which the atom is stabilized by the strong laser field. It was shown in Ref. [31] that the coupling of the field-free atomic bound state with the continuum resulting from absorbing photons is linearly

proportional to a Bessel function with the quiver length α_0 as an argument. This phenomenon results in a "breathing" above-threshold-ionization (ATI) spectra [32]. When the laser frequency is much larger than the atomic frequency, then the time-averaging approximation is applicable and the KH potential given above in Eqs. (10) and (11) is the leading term in the Fourier expansion of the time-dependent potential obtained within the framework of the acceleration gauge representation. As the laser frequency is increased the KH potential becomes more dominant in the Fourier expansion and the photoionization decay rate Γ is exponentially suppressed. This behavior is expected also for the photoionization decay rate of the core electron (this expectation will be checked by our illustrative numerical studies presented below). The question we address ourselves here is, how does the core electron decay rate depend on nuclear charge as a function of laser frequency?

When the duration of the laser pulse is sufficiently long the photoinduced dynamics is described by the Floquet Hamiltonian where $f(t)$ and therefore $g(\tau)$ as defined above are taken as unity [30]. Within the framework of the length gauge representation the scaled width Γ_{scaled} is associated with the imaginary part of the complex eigenvalue of the Floquet operator when outgoing boundary conditions are imposed on the Floquet solutions [29], i.e., $|\phi(\tau)\rangle = \exp[-i(E_{\text{scaled}} - i\Gamma_{\text{scaled}}/2)\tau/(\hbar\omega_{\text{scaled}})]|\varphi(\tau)\rangle$, where $\varphi(\tau) = \varphi(\tau + 2\pi n)$ is a periodic function and $n = 1, 2, \ldots$. The Floquet-Schrödinger equation after $\mathcal{Z}$ scaling is given by

$$\left[-i\hbar\omega_{\text{scaled}}\frac{\partial}{\partial\tau} - \frac{\hbar^2}{2m}\nabla^2_{x,y,z} - \frac{e^2}{\sqrt{x^2+y^2+z^2}}\right.$$
$$\left. + e\varepsilon_0^{\text{scaled}}\, z\, g(\tau)\cos(\tau)\right]|\varphi(\tau)\rangle$$
$$= \left[E_{\text{scaled}} - i\frac{\Gamma_{\text{scaled}}}{2}\right]|\varphi(\tau)\rangle \qquad (13)$$

with

$$E_{\text{scaled}} = \frac{E_r}{\mathcal{Z}^2}, \qquad (14)$$

$$\Gamma_{\text{scaled}} = \frac{\Gamma}{\mathcal{Z}^2}, \qquad (15)$$

where E_r and Γ are, respectively, the energy position and width (inverse lifetime) of the metastable (resonance) state. The last equation is exactly as obtained by Madsen and Lambropoulos [see Eq. (10) in Ref. [24]]. Here we get to a simple yet important point in our analysis. The photoionization rate of decay associated with the first peak in the ATI spectrum is linearly proportional to the laser intensity I_0 (see, for example, the calculation of the resonance width by applying the Fermi golden rule as given in the solution to problem 3.2 in Chap. 3 in Ref. [29]). Hence $\Gamma_{\text{H}} \propto I_0$, where Γ_{H} is the photoionization decay rate of the ground-state hydrogen atom for which $\mathcal{Z} = 1$. Therefore, we expect that in our case

$$\frac{\Gamma_{\text{scaled}}}{I_0^{\text{scaled}}} = \frac{\Gamma_{\text{H}}}{I_0}. \qquad (16)$$

Since $I_0 \propto \varepsilon_0^2$, we obtain

$$\Gamma_{\text{scaled}} = \left(\frac{\varepsilon_0^{\text{scaled}}}{\varepsilon_0}\right)^2 \Gamma_{\text{H}}. \qquad (17)$$

Using Eq. (8) and Eq. (15), we get that

$$\Gamma_{\text{H}} = \Gamma\mathcal{Z}^4. \qquad (18)$$

This simple scaling formula shows that for the same laser intensity (and not for the scaled one) the behavior of a one-electron system with $\mathcal{Z} > 1$ in a strong laser field of scaled frequency (7) is like that of a hydrogen atom. In the context of a many-electron atom, the ratio between the decay rate of the electron in the most diffused orbital and the decay rate of the electron in the $1s$ orbital is approximately equal to $\mathcal{Z}^4$.

The above result (18) has been verified by our illustrative numerical *ab initio* calculations. The values of Γ have been computed based on a variational Ritz method by applying the complex scaling transformation [29] and by representing the wave function by 11 Fourier basis functions, $\{\exp(i\omega n t)\}_{n=0,\pm1,\ldots,\pm5}$. As spatial basis functions 480 Slater-type orbitals (STOs) of $L = 0, \ldots, 5$ and $M = 0$ have been used. For each value of L, we have taken 80 STOs. Since the the symmetry is axial, M is a good quantum number. In our computations we have considered $M = 0$ states only. Such constructed basis set has proved to obtain converged results. The field amplitude was held at $\varepsilon_0 = 0.025$ a.u. For more details of calculations see Ref. [33]. Since the $1s$ core electron and the valence electron are approximately described by a one-electron ion ($\mathcal{Z} > 1$) and a hydrogen atom ($\mathcal{Z} = 1$), respectively, we plot in Fig. 1 the variation of Γ as a function of the laser frequency for hydrogenlike atoms for $\mathcal{Z} = 1, 2, 3, 4$. In turn, in Fig. 2 we present that $\Gamma\mathcal{Z}^4$ vs $\omega_{\text{scaled}} = \omega/\mathcal{Z}^2$ has a $\mathcal{Z}$-independent functional behavior. It clearly shows, in the context of the condition for the applicability of the KHA, that the laser frequency required to stabilize the core electron in the $1s$ orbital is larger by the factor $\mathcal{Z}^2$ than the laser frequency required to stabilize the electron in the most diffused orbital. Unfortunately, it is still difficult to specify the mutual relations between all the above quantities. However, in general one can write

$$\mathcal{Z}^4\Gamma(\varepsilon_0, \omega/\mathcal{Z}^2) = F(\varepsilon_0, \omega), \qquad (19)$$

where F does not depend on the atomic number. The fact that F is a nonlinear function indicates the importance of our results presented in Figs. 1 and 2.

A better estimate of the condition for the laser frequency for which the Kramers-Henneberger approximation is applicable for the core electrons can be obtained by carrying out time-independent perturbational analysis. In such analysis the KH dressed Hamiltonian in Eq. (10) is the zero-order Hamiltonian (see Refs. [34,35]). Let us assume that the calculations for a hydrogen atom show that at a given laser intensity and for $\omega \geqslant \omega_{\text{H}}$ the Hamiltonian (10) is indeed the dominant term (the higher-order terms can be neglected) in the perturbation series expansion of the exact solution of the Schrödinger equation as defined in Refs. [34,35]. In order to keep the same condition for the applicability of the KHA obtained for the hydrogen atom also for the core electron we should require the laser
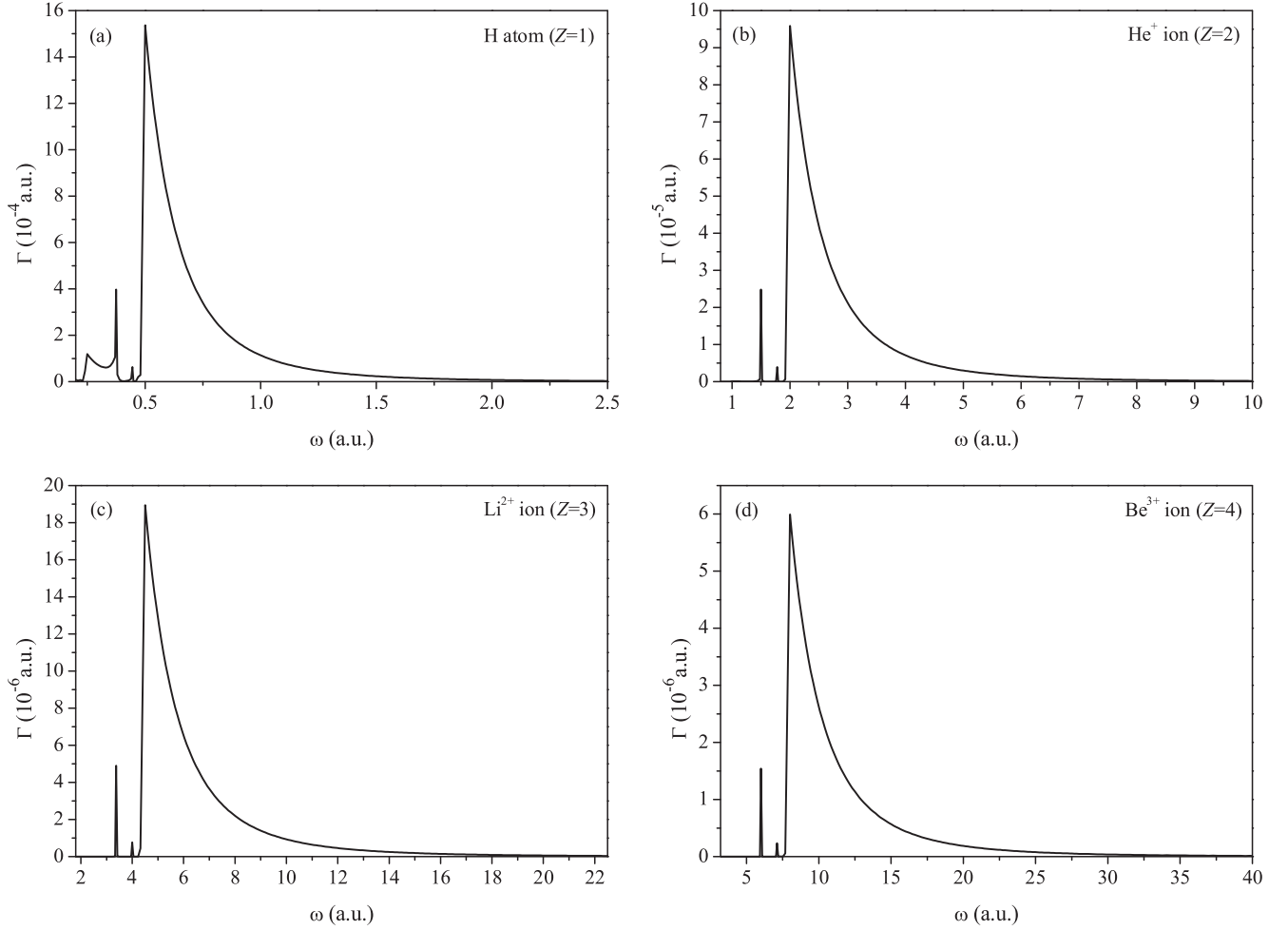
FIG. 1. The width of the resonance state associated with the ground state of a (a) H atom ($\mathcal{Z} = 1$), (b) He$^+$ ion ($\mathcal{Z} = 2$), (c) Li^{2+} ion ($\mathcal{Z} = 3$), and (d) Be^{3+} ion ($\mathcal{Z} = 4$) as a function of the laser frequency. The laser field amplitude is held at $\varepsilon_0 = 0.025$ a.u.

frequency to satisfy the following inequality:

$$\omega \geqslant \omega_{\mathrm{H}} \mathcal{Z}^2. \tag{20}$$

However, the results in Figs. 1 and 2 present that the photoionization decay rates are substantially reduced as $\mathcal{Z}$ is increased. The question arises: How much the condition given in Eq. (20) is too strong for the requirement of the applicability of the KHA for the core electron? In order to answer this question we plotted the decay rates Γ_{H}, Γ_{He^+}, $\Gamma_{\mathrm{Li}^{2+}}$, and $\Gamma_{\mathrm{Be}^{3+}}$ as a function of ω divided by $\mathcal{Z}$. The results illustrated in Fig. 3 clearly show that when H, He$^+$, Li^{2+}, and Be^{3+} ($\mathcal{Z} = 1,2,3,4$, respectively) interact with a laser field for a given intensity, then the high laser frequency which is required to have $\Gamma_{\mathrm{H}} = \Gamma_{\mathrm{He}^+} = \Gamma_{\mathrm{Li}^{2+}} = \Gamma_{\mathrm{Be}^{3+}}$ is about linearly proportional to $\mathcal{Z}$ and not to the square of atomic number as might be expected on the basis of the scaling laws derived above. A simple explanation is as follows. The field-free Hamiltonian for a hydrogenlike ion ($\mathcal{Z} > 1$) is given by

$$\hat{H}_{\mathrm{ion}} = -\frac{\hbar^2}{2m} \nabla_{\vec{r}}^2 - \frac{e^2}{|\vec{r}/\mathcal{Z}|}. \tag{21}$$

By carrying out the transformation $\vec{r} \rightarrow \vec{r}/\mathcal{Z}$ and $\hbar \rightarrow \hbar/\mathcal{Z}$, we obtain the Hamiltonian for a hydrogen atom. Using the scaling we shift the ground energy level of an ion to be as
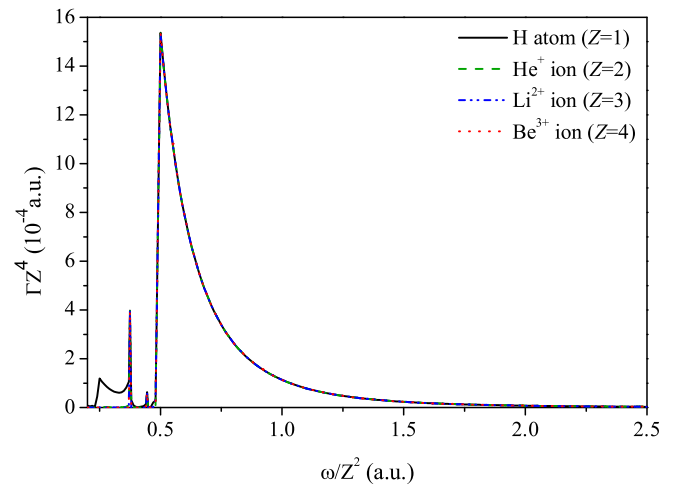

FIG. 2. (Color online) The width of the resonance state multiplied by $\mathcal{Z}^4$ associated with the ground state of a H atom ($\mathcal{Z} = 1$), He$^+$ ion ($\mathcal{Z} = 2$), Li^{2+} ion ($\mathcal{Z} = 3$), and Be^{3+} ion ($\mathcal{Z} = 4$) as a function of the laser frequency divided by $\mathcal{Z}^2$, where $\mathcal{Z}$ is the nuclear charge of the atom or ion. The laser field amplitude is held at $\varepsilon_0 = 0.025$ a.u.

FIG. 3. (Color online) The width of the resonance state associated with the ground state of a H atom ($\mathcal{Z} = 1$), He$^+$ ion ($\mathcal{Z} = 2$), Li^{2+} ion ($\mathcal{Z} = 3$), and Be^{3+} ion ($\mathcal{Z} = 4$) as a function of the laser frequency divided by the nuclear charge of the atom or ion. The laser field amplitude is held at $\varepsilon_0 = 0.025$ a.u.

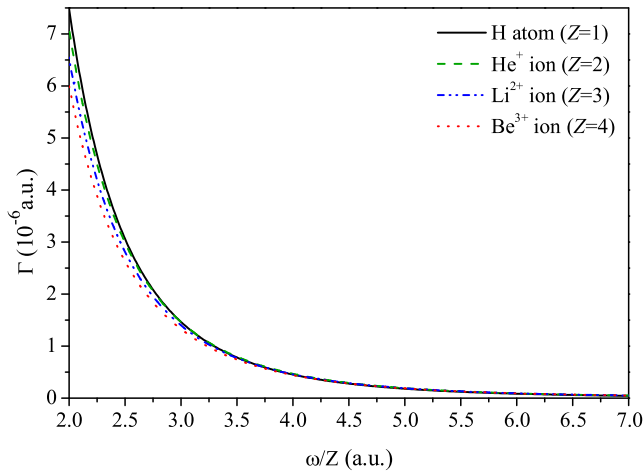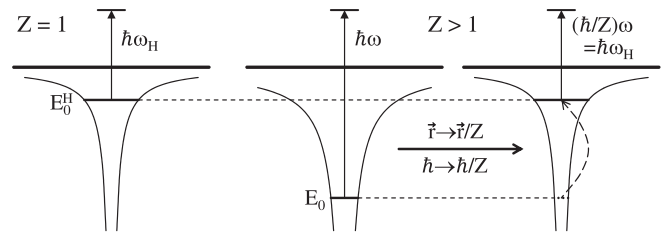FIG. 4. Schematic diagram of the photoionization due to the absorption of a single photon only for a hydrogen atom ($\mathcal{Z} = 1$) and a hydrogenlike ion ($\mathcal{Z} > 1$). After the transformation, $\vec{r} \rightarrow \vec{r}/\mathcal{Z}$ and $\hbar \rightarrow \hbar/\mathcal{Z}$, the energy level of the ground state of an ion is shifted up to the position corresponding to the energy level of the ground state of a hydrogen atom. Then, the decay rates of both systems are the same when $(\hbar/\mathcal{Z})\omega = \hbar\omega_\mathrm{H}$. Namely, for a given laser intensity the one-electron ion (which approximately describes the 1s core electron in a many-electron atom) and the hydrogen atom have about the same photoionization decay rate when $\omega = \omega_\mathrm{H}\mathcal{Z}$, provided this is a single-photon dynamical process.

the ground energy level of a hydrogen atom. It implies that the photoionization decay rate of a hydrogenic ion is the same as that of a hydrogen atom when $(\hbar/\mathcal{Z})\omega = \hbar\omega_\mathrm{H}$. Hence, we get $\omega = \omega_\mathrm{H}\mathcal{Z}$. This simple analytical result is in agreement with our numerical calculations. Note that this result has been obtained under the assumption that the one-electron atomic, molecular, or mesoscopic system is ionized due to the absorption of a single photon only (as, for example, by a free-electron laser) regardless of the polarization of the laser field. A schematic diagram presenting our simple explanation is shown in Fig. 4.

The conclusion of our paper is that the applicability of the KHA for core electrons in the 1s orbital of an atom can be determined on the basis of the knowledge of the laser parameters (intensity and frequency) for which the KHA holds for a hydrogen atom (see, for example, Fig. 1 in Ref. [14]). We have presented that for a given laser intensity the 1s core electron is stabilized when the laser frequency is larger than a value which is obtained from a product of the square of the atomic number $\mathcal{Z}^2$ and the laser frequency ω_H for which the KHA is applicable for a hydrogen atom in its ground

electronic state. The fact that the photoionization decay rate is decreased as $\mathcal{Z}$ is increased for a fixed intensity and a fixed frequency divided by $\mathcal{Z}^2$ shows that the condition we derived for stabilization is too strict. The softer condition is to find out for what value of R one gets that $\Gamma(\omega = \omega_\mathrm{H}R) = \Gamma_\mathrm{H}(\omega_\mathrm{H})$, where ω is the laser frequency required to stabilize the core electron of an atom with atomic number $\mathcal{Z}$. Based on the simple analytical arguments we have obtained that $R = \mathcal{Z}$. Our numerical calculations confirm this result. Thus, $\omega = \omega_\mathrm{H}\mathcal{Z}$ is a lower bound for the laser frequency which is needed to stabilize a many-electron atom in the presence of high-frequency strong laser fields.

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