

Ionization induced by the ponderomotive force in intense and high-frequency laser fields

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ABSTRACT

Atomic stabilization is a universal phenomenon that occurs when atoms interact with intense and high-frequency laser fields. In this work, we systematically study the influence of the ponderomotive (PM) force, present around the laser focus, on atomic stabilization. We show that the PM force could induce tunneling and even over-barrier ionization to the otherwise stabilized atoms. Such effect may outweigh the typical multiphoton ionization under moderate laser intensities. Our work highlights the importance of an improved treatment of atomic stabilization that includes the influence of the PM force.

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I. INTRODUCTION

The interaction of atoms with intense and high-frequency laser fields has given rise to a variety of topics and extensive investigations for decades.^{1–3} The atomic stabilization against ionization is one of the most interesting phenomena, in which the ionization probability decreases with increasing field intensity. Perfect interpretation has been made on this phenomenon by switching from the laboratory frame to an accelerated frame, called the Kramers–Henneberger (KH) frame,⁴ where the nucleus quivers under the laser field and forms a time-dependent potential. The high-frequency Floquet theory (HFFT)⁵ is the main theoretical framework to be applied on this topic. The lowest order of HFFT yields the prediction of adiabatic stabilization in the high-frequency regime, in which the time-dependent potential that the electron experiences becomes static by time-averaging the dynamic one, and the distorted potential was named “dressed” potential, depending only on the quiver amplitude α_0 of the electron.^{5–8} Atoms in states of such potential are called KH atoms, forming a relatively stable structure.^{9,10} In a linearly polarized laser field, the dressed potential can be viewed as a potential formed by a charged line segment, twice the length of α_0 , along the polarized direction, where the charge density peaks at the two end points of the segment. For large α_0 , the ground-state wave function under

such a potential splits into two non-overlapping parts, exhibiting a “dichotomy” structure,^{7,11} which even leads to a double-slit interference, similar to that in a diatomic molecule.¹² Several theoretical and experimental studies have been performed in attempting to confirm the existence of KH atoms.^{13–16}

Study on atomic stabilization has attracted considerable attention since its discovery. The past theoretical studies mainly focused on two forms of stabilization, namely, adiabatic stabilization and dynamic stabilization. The former is related to multiphoton ionization, initially derived from HFFT, where the ionization rate reduces with increasing laser intensity and frequency.^{10,17–19} The latter is usually studied by numerically solving the time-dependent Schrödinger equation (TDSE) that takes account the full laser–atom interaction, thereby establishing the relation between the survival probability of atoms and laser parameters.^{20–30} As for experimental studies, several experiments have also confirmed the existence of stabilization.^{31–34} However, certain factors may lead to the breakdown of atomic stabilization. For example, non-adiabatic effects brought by turn-on and turn-off of an ultra-short laser pulse may induce “shake-off” ionization;^{25,35,36} non-dipole and relativistic effects at high intensities may also contribute to destabilization.^{37–44} By far, the effect of ponderomotive (PM) force on atomic stabilization has not been studied. For a plane-wave field,

the PM energy is spatially constant and can be omitted as it should be. For a typical laser pulse, however, the intensity follows a Gaussian profile at the focus, where electrons will experience PM forces from the non-uniform spatial distribution of intensity.^{14,45–47} A recent study reveals that even a slight component of the PM force along the polarization direction can break the symmetry and change the electronic structure of KH atoms.⁴⁸ In this article, we investigate the effect of the PM force on atomic stabilization of KH atoms. We show that the PM force could induce tunneling and even over-barrier ionization to the otherwise stabilized atoms, thereby leading to the breakdown of stabilization.

This article is organized as follows: In Sec. II we detail the ionization rate induced by the PM force and discuss its dependence on system parameters. In Sec. III, we review the multiphoton ionization of KH atoms and show how the PM force influences its characteristics. In Sec. IV, we present a comparison of the PM-force-induced ionization and multiphoton ionization within a typical laser setting. Conclusions are given in Sec. V. Atomic units are used throughout unless stated otherwise.

II. PONDEROMOTIVE-FORCE-INDUCED TUNNELING IONIZATION AND OVER-BARRIER IONIZATION

A. Ponderomotive force and its effects on KH atoms

In the simplest case, we consider atoms with a single active electron (SAE). Under the SAE approximation, the TDSE in lab frame is written as

$$H_L \Psi_L = \left\{ \left(\mathbf{p} + \frac{1}{c} \mathbf{A}(\mathbf{r}, t) \right)^2 + V(\mathbf{r}) \right\} \Psi_L = i \partial_t \Psi_L, \quad (1)$$

where \mathbf{p} is the momentum, $\mathbf{A}(\mathbf{r}, t)$ is the vector potential of the laser field, and $V(\mathbf{r})$ is the Coulomb potential of the nucleus. Applying the KH transformation

$$U_{KH} = \exp [i \boldsymbol{\alpha}(\mathbf{r}, t) \cdot \mathbf{p}], \quad (2)$$

we transform the reference frame from the laboratory frame to an accelerated frame, called the KH frame, where

$$\boldsymbol{\alpha}(\mathbf{r}, t) = \frac{1}{c} \int^t \mathbf{A}(\mathbf{r}, \tau) d\tau \quad (3)$$

describes the quiver motion of a classical electron in the laser field. The transformed wave function reads

$$\Psi_{KH}(\mathbf{r}, t) = U_{KH} \Psi_L(\mathbf{r}, t) = \Psi_L(\mathbf{r} - \boldsymbol{\alpha}(\mathbf{r}, t), t). \quad (4)$$

The Hamiltonian, after neglecting higher order terms,⁴¹ is rewritten as

$$H_{KH} = \frac{\mathbf{p}^2}{2} + V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t)) + \frac{\mathbf{A}^2(\mathbf{r}, t)}{2c^2}, \quad (5)$$

where the first two terms define the KH atom we are familiar with.

Now, we consider the last term of Eq. (5), namely, $\mathbf{A}^2(\mathbf{r}, t)/2c^2$. For a laser pulse, linearly polarized in x direction and propagating along z direction, the vector potential is expressed as

$$\mathbf{A}(\mathbf{r}, t) = \frac{c}{\omega} \mathcal{E}_0(\mathbf{r}, t) \sin(\omega t - kz) \hat{\mathbf{x}}, \quad (6)$$

with $\mathcal{E}_0(\mathbf{r}, t)$ the electric field amplitude, ω the laser frequency, k the wave vector, and $\hat{\mathbf{x}}$ the polarization direction. Taking the cycle-average of $\mathbf{A}^2(\mathbf{r}, t)/2c^2$ gives rise to the PM potential

$$H_{PM} = \left\langle \frac{\mathbf{A}^2(\mathbf{r}, t)}{2c^2} \right\rangle = |\mathcal{E}_0(\mathbf{r}, t)|^2 / 4\omega^2. \quad (7)$$

For a plane wave, H_{PM} is constant everywhere, and, therefore, can be ignored. For a typical focused laser beam, however, the non-uniform spatial distribution of the PM potential at the laser focus results in the PM force

$$\mathbf{F}_{PM} = -\nabla H_{PM} = -\frac{1}{4\omega^2} \nabla |\mathcal{E}_0(\mathbf{r}, t)|^2. \quad (8)$$

To present an example, we assume a linearly polarized laser beam with Gaussian spatial intensity distribution, propagating along the z axis, which can be specified in cylindrical coordinates,

$$I(\mathbf{r}, t) = |\mathcal{E}_0(\mathbf{r}, t)|^2 = I_0 \left(\frac{w_0}{w(z)} \right)^2 \exp \left(-\frac{2r^2}{w(z)^2} \right) f(\eta), \quad (9)$$

where I_0 is the peak intensity and $w(z) = w_0 \sqrt{1 + (z/z_R)^2}$, with w_0 the beam waist and $z_R = \pi w_0^2 / \lambda$ the Rayleigh length. $f(\eta) = \exp[-(\eta/c\tau)^2]$, with $\eta = ct - z$ a Gaussian laser pulse envelope profile of optical intensity with a finite pulse duration $\tau = \tau_{FWHM} / 2\sqrt{\ln 2}$, where τ_{FWHM} is the full width at half maximum of the laser pulse. For typical lasers, z_R is more than 100 times larger than w_0 , which implies that the gradient of the laser field and the corresponding PM force in the propagation direction are much smaller than those in the radial direction, and, thus, negligible.⁴⁵

In this article, we take the hydrogen atom as the simplest example. According to HFFT, in the high-frequency limit, the system can be viewed as quasi-stationary, and the dynamics of the electron can be illustrated by the stationary Schrödinger equation

$$\left[\frac{\mathbf{p}^2}{2} + V_0(\alpha_0; \mathbf{r}) - \mathbf{F}_{PM} \cdot \mathbf{r} \right] \Psi = E \Psi, \quad (10)$$

where $V_0(\alpha_0; \mathbf{r})$ is the dressed potential, which is defined as the time average of the oscillating potential $V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t))$:

$$V_0(\alpha_0; \mathbf{r}) = \langle V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t)) \rangle = -\frac{1}{2\pi} \int_0^{2\pi/\omega} \frac{dt}{|\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t)|}. \quad (11)$$

Designating $\boldsymbol{\alpha}(\mathbf{r}, t) = \alpha_0 \cos \omega t \hat{\mathbf{x}}$, with the quiver amplitude $\alpha_0 = |\mathcal{E}_0(\mathbf{r}, t)| / \omega^2$, the potential is expressed as

$$V_0(\alpha_0; \mathbf{r}) = -\frac{2}{\pi} \frac{1}{\sqrt{r_+ r_-}} K \left(\sqrt{(1 - \hat{\mathbf{r}}_+ \cdot \hat{\mathbf{r}}_-) / 2} \right), \quad (12)$$

where $\mathbf{r}_\pm = \mathbf{r} \mp \alpha_0 \hat{\mathbf{x}}$ are the position vectors relative to the two end points of the quiver motion, and $K(k)$ is the complete elliptic integral of the first kind. Since we are interested in situations where $\alpha_0 \sim 10^2$ a.u., which is a quiver amplitude large enough that even for visible light, the high frequency requirement $\omega \gg I_p$ (I_p is the ionization potential) is well satisfied.

Despite the fact that PM forces are usually much weaker compared to those from the laser field, they could have substantial

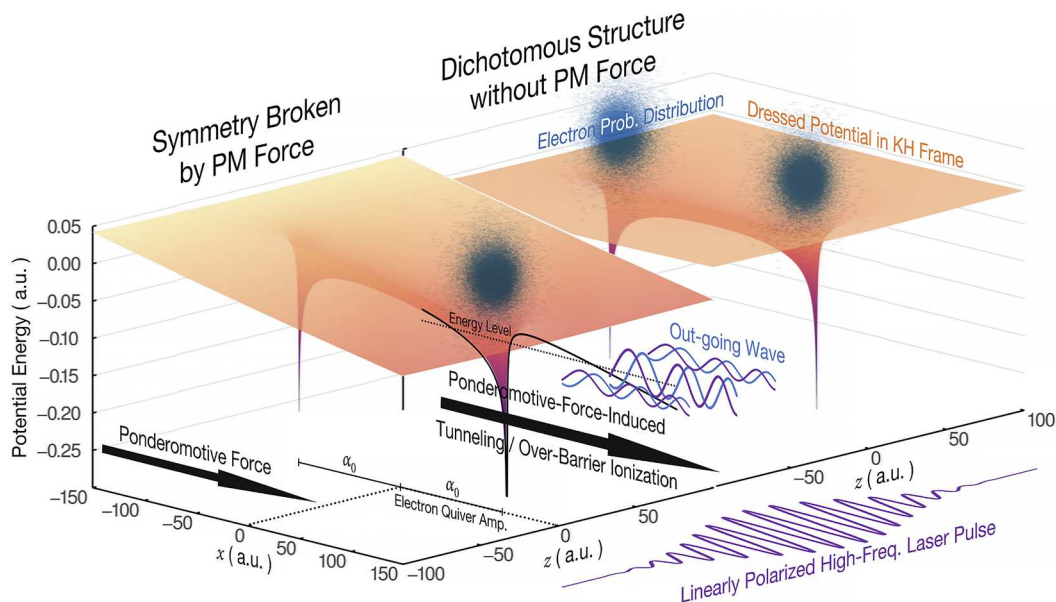


FIG. 1. Effect of the ponderomotive (PM) force on the structure of the KH atom in a linearly polarized laser field. The PM force may break the symmetric structure of the KH atom and further lead to tunneling ionization or even over-barrier ionization.

influence on the KH atoms' structure. The component of the PM force along the polarization direction will break the symmetric dichotomous structure of the KH atom, where the electron tends to stay near the “downstream” end point, exhibiting a single-lobe structure. Under the circumstances where the PM force is relatively strong, it would bend the KH potential, forming a potential barrier, which the electron may go across and escape to the continuum, resulting in tunneling ionization or over-barrier ionization. Figure 1 presents a schematic sketch of the effects of the PM force on the KH atom.

B. Ionization rate

In order to obtain the stationary ionization rate of KH atoms induced by the PM force, we implement a numerical solution of the TDSE.

In our scheme, the system evolves under the TDSE

$$i\partial_t\Psi = \left[-\frac{1}{2}\nabla^2 + V_0(\alpha_0; \mathbf{r}) - \Lambda(t)\mathbf{F}_{PM} \cdot \mathbf{r}\right]\Psi, \quad (13)$$

where a smooth turn on

$$\Lambda(t) = \begin{cases} 0, & t < 0, \\ \sin^2 \pi t/2T, & 0 \leq t \leq T, \\ 1, & t > T, \end{cases} \quad (14)$$

is implemented to ensure that the system goes through adiabatic transition from the field-free ($F_{PM} = 0$) initial state to the final state of stable ionization under the designated PM force. The initial state

is the ground eigenstate (1s state) of the field-free KH Hamiltonian, obtained through imaginary-time evolution. It is noteworthy that the purpose of our scheme is to obtain the stationary ionization rate induced by the PM force and its dependence on α_0 , F_{PM} , and θ . Thus, Eq. (13) is not used to mimic the exact physical dynamics of the KH atoms, but to calculate the stationary ionization rate induced by the PM force. The TDSE simulation is carried out on a three-dimensional Cartesian grid, and calculation of the initial state and propagation of the wave function are carried out using the split-operator Fourier method.⁴⁹ To avoid unphysical reflection on the boundaries, absorbing boundaries of the $\cos^{1/6}$ shape are applied after each time step propagation to suppress the out-going wave approaching the grid border. The ionization rate is defined as the surface integral of the probability current density

$$\Gamma(t) = \int_{\Sigma} \mathbf{J} \cdot d\mathbf{S} = \int_{\Sigma} \frac{i}{2} (\Psi \nabla \Psi^* - \Psi^* \nabla \Psi) \cdot d\mathbf{S}. \quad (15)$$

Soon after $\Lambda(t)$ reaches 1 and the system stabilizes, we have $\Gamma(t) \propto \exp(-\Gamma_0 t)$, thus, the stationary ionization rate can be derived from

$$\Gamma_0 = -\frac{d \ln \Gamma(t)}{dt}. \quad (16)$$

Figure 2 presents the static ionization rates obtained by numerically solving the TDSE [Eq. (13)] following the above procedure, where Fig. 2(a) is a sketch of the geometry of the problem.

As displayed in Figs. 2(b)–2(d), the F_{PM} -dependence of the ionization rate is obvious: For fixed α_0 , the ionization rate increases exponentially as F_{PM} increases since the potential barrier gets thinner and lower for larger F_{PM} . On the other hand, for a certain laser frequency, the PM-free, ground-state ionization potential $I_p = |E_0|$

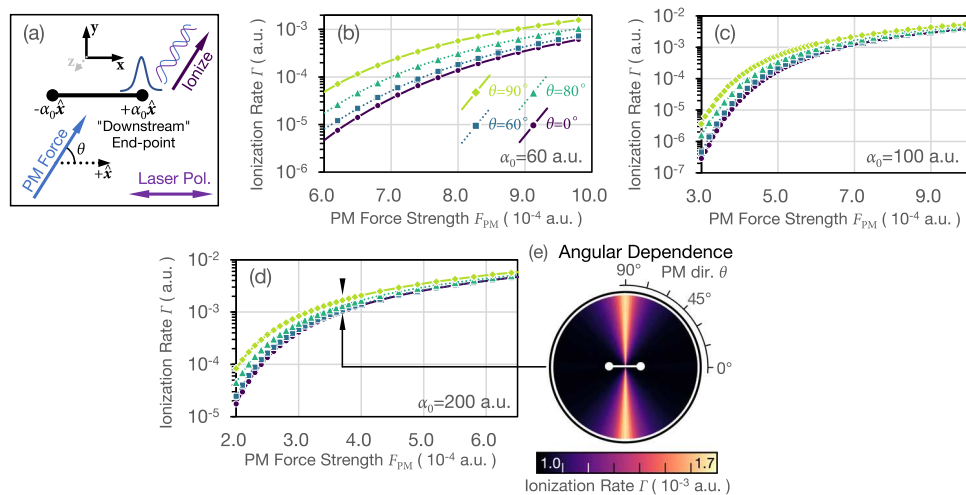


FIG. 2. PM-force-induced ionization rates, obtained by numerically solving the TDSE. (a) The x axis denotes the laser's polarization direction, and θ ($0^\circ \leq \theta \leq 90^\circ$) represents the angle between the PM force and the $+x$ direction. For $\theta \neq 90^\circ$, the end point $+\alpha_0 \hat{x}$ is the “downstream” one, i.e., $V_0(+\alpha_0 \hat{x}) < V_0(-\alpha_0 \hat{x})$. In most situations, the electron wave function concentrates near the “downstream” end point. The dependence of the ionization rate on F_{PM} and θ are presented in panel (b) with $\alpha_0 = 60$ a.u., panel (c) with $\alpha_0 = 100$ a.u., and panel (d) with $\alpha_0 = 200$ a.u. The angular dependence of the ionization rate under the parameter $\alpha_0 = 200$ a.u. and $F_{\text{PM}} = 3.7 \times 10^{-4}$ a.u. is presented in panel (e).

decreases and the PM force increases dramatically with increasing α_0 ; the PM-force-induced ionization is, therefore, much more common for larger α_0 . Since $\alpha_0 \propto \sqrt{I}$ and $F_{\text{PM}} \propto I$ (I is the laser intensity), the ionization induced by the PM force would rapidly grow as the laser intensity increases, eventually turning into a dominant factor that determines the lifetime of the KH atom.

From Fig. 2(e), we could further get a glimpse of the dependence of the ionization rate on the direction of the PM force: The ionization rate peaks when the PM force is perpendicular to the laser polarization direction and decreases as the PM force rotates toward the polarization direction. This trend can be understood when we look closely into the potential near the end points. As will be shown in Eq. (19) in Sec. II C, the angular dependence of the potential near the end points follows as

$$\tilde{V}_0 \propto -K \left(\sin \frac{\theta}{2} \right), \quad (17)$$

where θ is the angle between \mathbf{r} and \hat{x} . Such an angular dependence indicates that the potential drops monotonously as θ increases from 0° to 90° . Therefore, the PM force bends the potential more along $\theta = 90^\circ$ compared to $\theta = 0^\circ$, allowing larger out-flowing probability current. Another character of the angular dependence follows straightforwardly: The ratio between the ionization rates along $\theta = 90^\circ$ and $\theta = 0^\circ$ decreases as F_{PM} increases, which is in line with the expectation that ionization in weaker fields is more sensitive to the barrier thickness.

C. Large- α_0 and large- F_{PM} limits: An approximation using end-point scaling

We note that for sufficiently large α_0 , the eigenstate of the field-free KH Hamiltonian exhibits a dichotomous structure: The wave

function concentrates near the two end points, forming two non-overlapping parts. Hence, our interested region is in the vicinity of the end point $+\alpha_0 \hat{x}$ or $-\alpha_0 \hat{x}$, where $r_+ \ll \alpha_0$ or $r_- \ll \alpha_0$ is satisfied, and V_0 reduces to Ref. 11:

$$\begin{aligned} V_0(\alpha_0; \mathbf{r}) &\simeq \tilde{V}_0(\alpha_0; \mathbf{r}_+) & \text{for } r_+ \ll \alpha_0, \\ V_0(\alpha_0; \mathbf{r}) &\simeq \tilde{V}_0(\alpha_0; -\mathbf{r}_-) & \text{for } r_- \ll \alpha_0. \end{aligned} \quad (18)$$

The “end-point approximate” potential \tilde{V}_0 , is expressed as

$$\tilde{V}_0(\alpha_0; \mathbf{r}) = -\frac{2}{\pi} \frac{1}{\sqrt{2\alpha_0 r}} K \left(\sqrt{\frac{1 - \hat{\mathbf{r}} \cdot \hat{\mathbf{x}}}{2}} \right). \quad (19)$$

Under large- α_0 limit, the “end-point approximate” dynamics can be further scaled to obtain α_0 -independent results. Taking the end point $+\alpha_0 \hat{x}$ as an example, by introducing a scaled coordinate variable $\boldsymbol{\rho} = \alpha_0^{-1/3} \mathbf{r}_+$, the stationary Schrödinger equation [Eq. (10)] becomes

$$\left[-\frac{1}{2} \nabla_{\boldsymbol{\rho}}^2 + \tilde{V}'_0(\boldsymbol{\rho}) - \mathbf{F}'_{\text{PM}} \cdot \boldsymbol{\rho} \right] \Phi = W \Phi, \quad (20)$$

where

$$\tilde{V}'_0(\boldsymbol{\rho}) = -\frac{1}{\pi} \sqrt{\frac{2}{\rho}} K \left(\sqrt{\frac{1 - \hat{\boldsymbol{\rho}} \cdot \hat{\mathbf{x}}}{2}} \right) \quad (21)$$

is the scaled approximate potential, $W = \alpha_0^{2/3} E$ is the scaled energy, $\mathbf{F}'_{\text{PM}} = \alpha_0 \mathbf{F}_{\text{PM}}$ is the scaled PM force, and $\Phi(\boldsymbol{\rho}) = \alpha_0^{1/2} \Psi(\mathbf{r}_+)$ is the scaled normalized wave function (note that the origin has shifted to end point $+\alpha_0 \hat{x}$). Furthermore, we introduce the scaled time variable $\tau = \alpha_0^{-2/3} t$, thus, the TDSE, Eq. (13), neglecting $\Lambda(t)$, is rewritten as

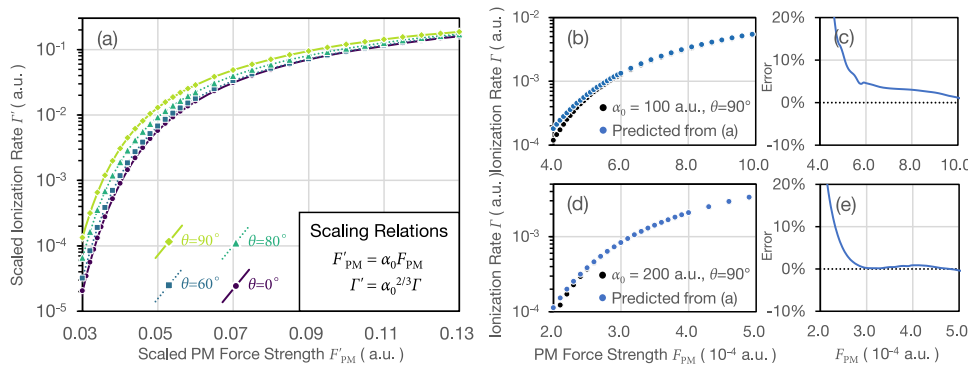


FIG. 3. (a) Ionization rates obtained through end-point scaling. (b) and (d) Comparison of actual ionization rates and those predicted from scaling, with corresponding errors shown in (c) and (e).

$$i\partial_t\Phi = \left[-\frac{1}{2}\nabla_\rho^2 + \tilde{V}'_0(\rho) - \mathbf{F}'_{PM} \cdot \rho \right] \Phi. \quad (22)$$

Equations (20) and (22) are both α_0 -independent. Finally, the ionization rate, which is of our interest, follows the scaling law

$$\Gamma' = \alpha_0^{2/3}\Gamma. \quad (23)$$

Figure 3(a) shows the scaled ionization rates obtained by solving the scaled TDSE [Eq. (22)] (smooth turn-on is also applied). This variable scaling scheme provides a way of presenting an α_0 -independent result, which can be used to predict physical quantities of the actual system of arbitrary α_0 as long as the end-point approximation is valid.

To examine the validity of our scaling scheme, we employ the data of the ionization rate of the scaled system to predict the ionization rate of the real system. The results for $\alpha_0 = 100$ a.u. and $\alpha_0 = 200$ a.u. at $\theta = 90^\circ$ are shown in Figs. 3(b) and 3(d), with corresponding errors shown in Figs. 3(c) and 3(e), respectively. It is apparent that the predicted ionization rates match well with the actual ones and match better for stronger PM forces. What is more, at a certain $F'_{PM} = \alpha_0 F_{PM}$, a larger α_0 gives a better match between predicted and actual ionization rates. To explain this, we refer to the end-point approximation that we made in the scaling scheme in Eq. (18), where we assumed the condition $r_+ \ll \alpha_0$ or $r_- \ll \alpha_0$. We recast such a condition from two aspects. For one thing, this condition requires the wave function to concentrate in the vicinity of two end points, which is satisfied only for $\alpha_0 > 20$ a.u. (large- α_0 condition). For the other thing, this condition requires tunneling or over-barrier ionization to occur close to the end points, where “the location the ionization occurs” can be characterized using the location of the tunneling exit or barrier top, which is satisfied for large F_{PM} . Therefore, the present scaling scheme is applicable in the large- α_0 and large- F_{PM} limit.

III. IMPACT OF PONDEROMOTIVE FORCE ON MULTIPHOTON IONIZATION

Multiphoton ionization (MPI) plays a key role in adiabatic stabilization in intense and high-frequency laser fields.^{3,10,17,18,50} In this section, we give a brief review on the multiphoton processes under the framework of HFFT and study the impact of the PM force on it.

Let us start from the space-translated Schrödinger equation in the KH frame, where the Hamiltonian is given by Eq. (5). Under the PM-free circumstance, the TDSE is written as

$$\left[\frac{\mathbf{p}^2}{2} + V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t)) \right] \Psi = i\partial_t \Psi. \quad (24)$$

The potential term, $V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t))$, is periodic; thus, a Floquet–Fourier-form solution with quasi-energy E can be sought:³

$$\Psi(\mathbf{r}, t) = e^{-iEt} \sum_{n=-\infty}^{+\infty} \Psi_n(\mathbf{r}) e^{-in\omega t}. \quad (25)$$

We also Fourier-analyze the potential, giving

$$V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t)) = \sum_{n=-\infty}^{+\infty} V_n(\alpha_0; \mathbf{r}) e^{-in\omega t}, \quad (26)$$

and the Fourier coefficients

$$V_n(\alpha_0; \mathbf{r}) = \frac{1}{2\pi} \int_0^{2\pi/\omega} V(\mathbf{r} + \boldsymbol{\alpha}(\mathbf{r}, t)) e^{in\omega t} dt. \quad (27)$$

Substituting Eqs. (25) and (26) into Eq. (24) gives an infinite system of coupled differential equations for $\Psi_n(\mathbf{r})$. The potential $V(\mathbf{r})$ has a Coulomb tail, which requires the solution to behave asymptotically as³

$$\Psi(\mathbf{r}, t) \rightarrow \frac{1}{(2\pi)^{3/2}} e^{-iEt} \sum_{n=-\infty}^{+\infty} e^{-in\omega t} f_n(\alpha_0, \omega; \hat{\mathbf{r}}) e^{(ik_n r - \ln(2k_n r)/k_n)/r}, \quad (28)$$

when $r \rightarrow \infty$, and the wave vectors follow the relation

$$k_n = \begin{cases} \sqrt{2(E + n\omega)}, & \text{for } E + n\omega \geq 0, \\ i\sqrt{2|E + n\omega|}, & \text{for } E + n\omega < 0. \end{cases} \quad (29)$$

Thus, for closed channels, where $E + n\omega < 0$, $\Psi_n(\mathbf{r})$ exponentially damps and vanishes at infinity, while for open channels, where $E + n\omega \geq 0$, $\Psi_n(\mathbf{r})$ is an out-going spherical wave, and $f_n(\alpha_0, \omega; \hat{\mathbf{r}})$, therefore, represents the n -photon ionization amplitude, with the corresponding n -photon decay rate Γ_n given by

$$\Gamma_n = k_n \int |f_n(\alpha_0, \omega; \hat{r})|^2 d\hat{r}. \quad (30)$$

The total decay rate Γ_{MPI} is subsequently the sum of Γ_n over all open channels.

An iteration scheme proposed by Gavrilu and Kamiński⁵ can be applied to study the solution of Eq. (24). The zeroth order iteration gives the familiar “adiabatic stabilization,” with

$$\Psi_n(\mathbf{r}) = \Psi_0(\mathbf{r})\delta_{0n}, \quad (31)$$

where $\Psi_0(\mathbf{r})$ is the eigenstate of the dressed potential, $V_0(\alpha_0; \mathbf{r})$, as discussed above in Sec. II. The first-order iteration gives the expression of f_n :

$$f_n(\alpha_0, \omega; \hat{r}) = -\frac{1}{\sqrt{2\pi}} \langle \Psi_{k_n}^{(-)} | V_n | \Psi_0 \rangle, \quad (32)$$

where $|\Psi_{k_n}^{(\pm)}\rangle$ denotes the scattering solution of

$$\left[\frac{\mathbf{p}^2}{2} + V_0(\alpha_0; \mathbf{r}) \right] \Psi = (E_0 + n\omega) \Psi, \quad (33)$$

and behaves as a plane wave with wave vector \mathbf{k}_n (with the same direction as \hat{r}) plus an outgoing (+) or incoming (−) spherical wave, which are respectively normalized to the delta function in the momentum space. It’s also worth noting that in order for the iteration to converge and the first-order iteration to represent a good approximation, the frequency of the laser needs to be sufficiently large, and more explicitly, $\omega \gg I_p = |E_0|$ is needed, where E_0 is the ground-state energy of the potential V_0 .

In order to obtain the decay rate of MPI, we apply Eq. (32), and replace $|\Psi_{k_n}^{(-)}\rangle$ with a plane wave $|k_n\rangle$, which is, in fact, the first-order Born approximation. The calculation is still carried out numerically on a Cartesian grid. Under PM-free circumstances, Ψ_0 is obtained by simply using an imaginary-time evolution, while for those with the PM force, as the eigenstate exhibits a continuum character beyond the downstream end point, the imaginary-time evolution is unable to give a ground state. Under such circumstances, we use a trick of placing a wide absorbing boundary in the downstream direction to suppress probability growth in the continuum region during the evolution. By properly setting the parameter of the absorbing boundary, the imaginary-time evolution gives a satisfactory bound eigenstate, which exhibits a single-lobe structure.

Shown in Fig. 4 are the angle-dependent MPI rates $k_n |f(\hat{r})|^2$, obtained with the above scheme. The total ionization rate is a little influenced by the presence of the PM force, while the angular rate, which corresponds to the photoelectron momentum distribution, has shown substantial structure alteration under the PM force. Clearly, the PM-free case with an initial state that has a dichotomous structure exhibits interference fringes in the angular rates. It was interpreted by Pont as resulting from the interference of electron waves ejected from two end points.¹⁸ However, the component of the PM force along the polarization direction results in a single-lobe structure in the initial state, effectively closing one of the slits, thereby eliminating the fringes. It is found that even a tiny PM force along the polarization direction is capable of breaking the dichotomy; we can infer that for a common laser frequency

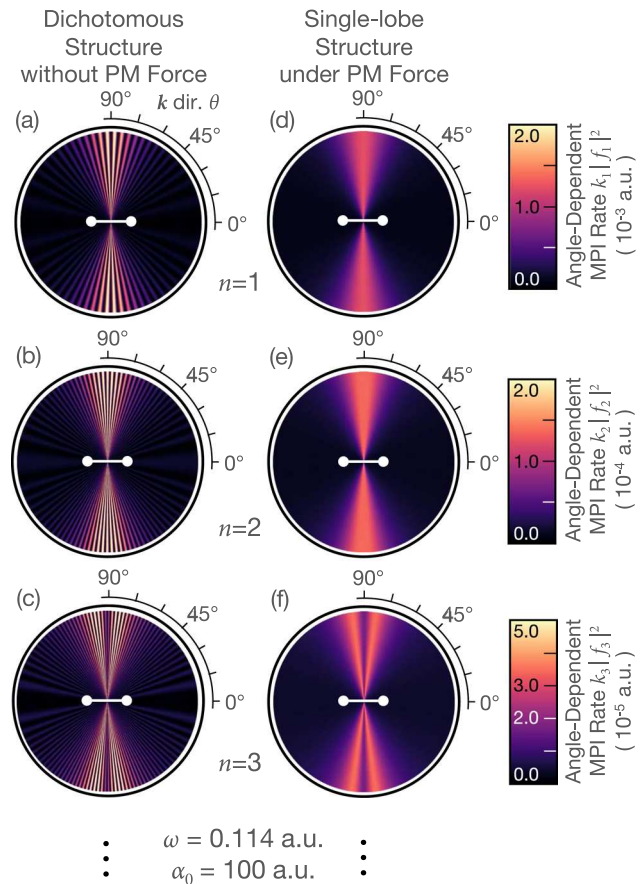


FIG. 4. Angle-dependent multiphoton ionization rates $k_n |f(\hat{r})|^2$ from (a)–(c), PM-free, and (d)–(f), PM-existing ground states, with $\omega = 0.114$ a.u. and $\alpha_0 = 100$ a.u. The first, second, and third rows display the single-, double-, and triple-photon ionization rates, respectively.

(visible light or UV) and high laser intensities ($\alpha_0 > 50$ a.u.), a little portion of the KH atoms can preserve their dichotomy geometry and contribute to the interference signal in a real laser field.

IV. PONDEROMOTIVE-FORCE-INDUCED TUNNELING/OVER-BARRIER IONIZATION VS MULTIPHOTON IONIZATION

In this section, we take the Gaussian laser of Eq. (9) as an example. We assign the laser parameter $\lambda = 400$ nm ($\omega = 0.114$ a.u.), $w_0 = 17.5$ μm , with two peak intensities, $I_0 = 2 \times 10^{17}$ W/cm^2 and $I_0 = 3 \times 10^{17}$ W/cm^2 . All the laser parameters, as well as PM forces used in the calculations, are accessible in experiments. We note that such laser parameters have already been achieved.⁵¹ For atoms in the focal plane ($z = 0$) within one beam waist ($r \leq w_0$), the high-frequency criteria are well satisfied. Figure 5 displays the comparison of ionization rates of PM-force-induced ionization and MPI at different locations on the focal plane. For a radial intensity distribution of Gaussian type, the PM force is pointing outward along the radial direction, with a strength increasing from zero to maxima as the

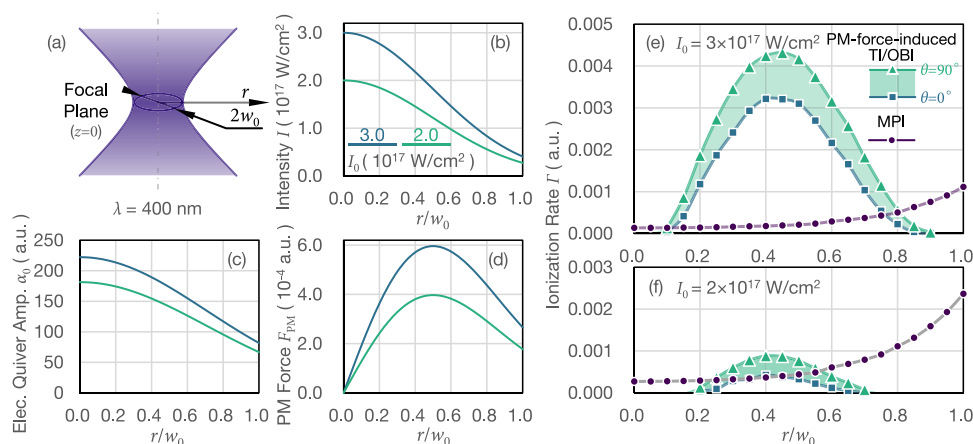


FIG. 5. A comparison of the PM-force-induced tunneling ionization (TI)/over-barrier ionization (OBI) and multiphoton ionization (MPI) rates on the focal plane of a linearly polarized Gaussian beam. (a) The laser has a wavelength of $\lambda = 400$ nm, w_0 is the beam waist, and r denotes the radial distance from the focal axis. Two peak intensities, $I_0 = 2 \times 10^{17}$ W/cm² and $I_0 = 3 \times 10^{17}$ W/cm², are used. (b)–(d) display the distribution of intensity, the electron quiver amplitude, and the strength of the PM force on the focal plane, respectively. (e) and (f) show the comparison between the PM-force-induced TI/OBI and MPI rates of lasers, with two given peak laser intensities.

radial location moves from zero to half beam waist ($r/w_0 = 0.5$), and slowly decaying when moving further out [Fig. 5(d)]. Although the PM force peaks at half beam waist, the ionization induced by the PM force peaks a bit inward, as is clear from Figs. 5(e) and 5(f). The reason is that the laser intensity itself is greater near the beam axis, resulting in a larger α_0 and lower I_p , thereby effectively shifting the peak of the PM-force-induced ionization to be inside the location of the half beam waist. Comparison of Figs. 5(e) and 5(f) also reveals that as the intensity increases, MPI gets restrained due to the increase of α_0 , while the PM-force-induced ionization grows exponentially and takes over as the dominant factor contributing to ionization, resulting in destabilization.

V. CONCLUSIONS

Atomic stabilization in intense and high-frequency laser fields has attracted widespread attention and discussion for decades, and is convinced to result in decreasing ionization probability with increasing laser intensity. However, the present work indicates that as the laser intensity increases, the PM force, which is induced by the non-uniform spatial distribution of laser intensity near the focal spot, will not only break the symmetry of the well-known dichotomous structure, but could also bring about tunneling and even over-barrier ionization, resulting in a breakdown of atomic stabilization. Our work casts light on the importance of an improved model of atomic stabilization that accounts for the influence of the PM force.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Mingyu Zhu: Methodology (lead); Software (lead); Visualization (lead); Writing – original draft (lead); Writing – review & editing (equal). **Yuxiang Liu:** Methodology (equal); Software (equal). **Chunli Wei:** Methodology (equal); Software (equal). **Hongcheng Ni:** Conceptualization (equal); Investigation (equal); Methodology (equal); Software (equal); Writing – review & editing (equal). **Qi Wei:** Conceptualization (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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