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Kaoru Yamanouchi
Katsumi Midorikawa
Luis Roso *Editors*

Progress in Ultrafast Intense Laser Science XVI

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Kaoru Yamanouchi · Katsumi Midorikawa ·
Luis Roso
Editors

Progress in Ultrafast Intense Laser Science XVI

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Editors
Kaoru Yamanouchi
Department of Chemistry
School of Science, The University of Tokyo
Tokyo, Japan

Katsumi Midorikawa
Center for Advanced Photonics
RIKEN, Wako, Saitama, Japan

Luis Roso
Centro de Laseres Pulsados
Vega de Salamanca, Salamanca, Spain

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Preface

We are pleased to present the sixteenth volume of Progress in Ultrafast Intense Laser Science. As the frontiers of ultrafast intense laser science rapidly expand ever outward, there continues to be a growing demand for an introduction to this interdisciplinary research field that is at once widely accessible and capable of delivering cutting-edge developments. Our series aims to respond to this call by providing a compilation of concise review-style articles written by researchers at the forefront of this research field, so that researchers with different backgrounds as well as graduate students can easily grasp the essential aspects.

As in the previous volumes, each chapter of this book begins with an introductory part, in which a clear and concise overview of the topic and its significance is given, and moves onto a description of the authors' most recent research results. All chapters are peer-reviewed. The articles of this sixteenth volume cover a diverse range of the interdisciplinary research field, and the topics may be grouped into three categories: atoms and molecules in intense laser fields (Chaps. 1– 5), applications of circularly polarized laser pulses (Chaps. 6 and 7), and theoretical and technological developments for intense laser field experiments (Chaps. 8– 10).

From the third volume, the PUILS series has been edited in liaison with the activities of the Center for Ultrafast Intense Laser Science at the University of Tokyo, which has also been responsible for sponsoring the series and making the regular publication of its volumes possible. From the fifth volume, the Consortium on Education and Research on Advanced Laser Science, the University of Tokyo, has joined this publication activity as one of the sponsoring programs. The series, designed to stimulate interdisciplinary discussion at the forefront of ultrafast intense laser science, has also collaborated since its inception with the annual symposium series of ISUILS (http://www.isuils.jp/), sponsored by JILS (Japan Intense Light Field Science Society).

We would like to take this opportunity to thank all the authors who have kindly contributed to the PUILS series by describing their most recent work at the frontiers of ultrafast intense laser science. We also thank the reviewers who have read the submitted manuscripts carefully. One of the co-editors (KY) thanks Ms. Mihoshi Abe for her help with the editing processes.

We hope this volume will convey the excitement of ultrafast intense laser science to the readers and stimulate interdisciplinary interactions among researchers, thus paving the way to explorations of new frontiers.

Tokyo, Japan — Kaoru Yamanouchi
Saitama, Japan — Katsumi Midorikawa
Salamanca, Spain — Luis Roso
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Contributors

Hiroshi Akagi Kansai Photon Science Institute, National Institutes for Quantum and Radiological Science and Technology (QST), Kyoto, Japan

Toshiaki Ando Department of Chemistry, School of Science, The University of Tokyo, Tokyo, Japan

André D. Bandrauk Computational Chemistry & Molecular Photonics, Faculté des Sciences, Université de Sherbrooke,Laboratoire de Chimie Théorique, Sherbrooke, Canada

Junchi Chen State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Lingru Chen State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Farhad H. M. Faisal Fakultät Für Physik, Universität Bielefeld, Bielefeld, Germany

Robert Fedosejevs Electrical and Computer Engineering, University of Alberta, Edmonton, AB, Canada

Yao Fu State Key Laboratory of Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun, China

Zebiao Gan State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Jing Guo Institute of Atomic and Molecular Physics, Jilin University, Changchun, China

Eisuke Haraguchi Department of Applied Physics, Hokkaido University, Sapporo, Japan

Feng He Key Laboratory for Laser Plasmas (Ministry of Education) and School of Physics and Astronomy, Collaborative Innovation Center for IFSA (CICIFSA), Shanghai Jiao Tong University, Shanghai, China;
CAS Center for Excellence in Ultra-intense Laser Science, Shanghai, China

Pei-Lun He Key Laboratory for Laser Plasmas (Ministry of Education), School of Physics and Astronomy, Shanghai, China;
Collaborative Innovation Center for IFSA (CICIFSA), Shanghai Jiao Tong University, Shanghai, China

Ryuji Itakura Kansai Photon Science Institute, National Institutes for Quantum and Radiological Science and Technology (QST), Kyoto, Japan

Kengo Ito Department of Applied Physics, Hokkaido University, Sapporo, Japan

Atsushi Iwasaki Department of Chemistry, School of Science, The University of Tokyo, Tokyo, Japan

Keisuke Kaneshima Department of Applied Physics, Hokkaido University, Sapporo, Japan

Tsuyoshi Kato Department of Chemistry, School of Science, The University of Tokyo, Tokyo, Japan

Toshiyuki Kawashima Central Research Laboratory, Industrial Development Center, Hamamatsu Photonics K.K., Hamamatsu City, Shizuoka, Japan

Yosuke Kayanuma Laboratory for Materials and Structures, Tokyo Institute of Technology, Nagatsuta, Yokohama, Japan;
Department of Physical Sciences, Osaka Prefecture University, Sakai, Osaka, Japan

Hirohiko Kono Department of Chemistry, Graduate School of Science, Tohoku University, Sendai, Japan

Shiro Koseki Department of Chemistry, Graduate School of Science, Osaka Prefecture University, Osaka, Japan

Norio Kurita Central Research Laboratory, Industrial Development Center, Hamamatsu Photonics K.K., Hamamatsu City, Shizuoka, Japan

Yuxin Leng State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Roberto Lera Center for Pulsed Lasers, CLPU, Villamayor, Salamanca, Spain

Helong Li State Key Laboratory of Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun, China

Ruxin Li State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Shuai Li State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Wenqi Li State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Xiaoyan Liang State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Xinyan Liu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Yanqi Liu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Erik Lötstedt Department of Chemistry, School of Science, The University of Tokyo, Tokyo, Japan

Hideki Ohmura National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki, Japan

Shu Ohmura Department of Physical Science and Engineering, Nagoya Institute of Technology, Nagoya, Japan

Tomohito Otobe Kansai Photon Science Institute, National Institutes for Quantum and Radiological Science and Technology (QST), Kyoto, Japan

Yujie Peng State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

José Antonio Pérez-Hernández Center for Pulsed Lasers, CLPU, Villamayor, Salamanca, Spain

Luis Roso Center for Pulsed Lasers, CLPU, Villamayor, Salamanca, Spain; Applied Physics Department, Universidad de Salamanca, Salamanca, Spain

Taro Sekikawa Department of Applied Physics, Hokkaido University, Sapporo, Japan

Takashi Sekine Central Research Laboratory, Industrial Development Center, Hamamatsu Photonics K.K., Hamamatsu City, Shizuoka, Japan

Yunhai Tang State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Cheng Wang State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Siqi Wang State Key Laboratory of Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun, China

Xiaobin Wang State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Xinliang Wang State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Huailiang Xu State Key Laboratory of Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun, China;
CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai, China

Lu Xu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Yi Xu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Zhizhan Xu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Kaoru Yamanouchi Department of Chemistry, School of Science, The University of Tokyo, Tokyo, Japan

Bo Yao State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Dinjun Yin State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Lianghong Yu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Linpeng Yu State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Kai-Jun Yuan Institute of Atomic and Molecular Physics, Jilin University, Changchun, China;
Laboratoire de Chimie Théorique, Faculté des Sciences, Université de Sherbrooke, Québec, Sherbrooke, Canada

Xiaobo Zhang State Key Laboratory of High Field Laser Physics and CAS Center for Excellence in Ultra-Intense Laser Science, Shanghai Institute of Optics and Fine Mechanics(SIOM), Chinese Academy of Sciences(CAS), Shanghai, China

Zhao-Han Zhang Key Laboratory for Laser Plasmas (Ministry of Education), School of Physics and Astronomy, Shanghai, China;
Collaborative Innovation Center for IFSA (CICIFSA), Shanghai Jiao Tong University, Shanghai, China

Chapter 1
Robust Strategies for Affirming Kramers-Henneberger Atoms

Pei-Lun He, Zhao-Han Zhang, and Feng He

Abstract Atoms exposed to high-frequency strong laser fields experience ionization suppression due to the deformation of Kramers-Henneberger (KH) wave functions, which has not been confirmed yet in any experiment. We propose a bichromatic pump-probe strategy to affirm the existence of KH states, which are formed by the pump pulse and ionized by the probe pulse. In the case of the single-photon ionization triggered by a vacuum ultra-violet probe pulse, the double-slit character of the KH atom is mapped to the photoelectron momentum distribution. In the case of the tunneling ionization induced by an infrared probe pulse, streaking in anisotropic Coulomb potential gives rise to the rotation of the photoelectron momentum distribution in the laser polarization plane. Apart from bichromatic schemes, the non-Abelian geometric phase provides an alternative route to affirm the existence of KH states. Following specific loops in laser parameter space, a complete spin flipping transition could be achieved. Our proposal has the advantages of being robust against focal-intensity average as well as ionization depletion and is accessible with current laser facilities.

P.-L. He (✉) · Z.-H. Zhang
Key Laboratory for Laser Plasmas (Ministry of Education), School of Physics and Astronomy, Shanghai, China
e-mail: a225633@sjtu.edu.cn

Z.-H. Zhang
e-mail: zhangzhaohan@sjtu.edu.cn

Collaborative Innovation Center for IFSA (CICIFSA), Shanghai Jiao Tong University, Shanghai 200240, China

F. He
Key Laboratory for Laser Plasmas (Ministry of Education) and School of Physics and Astronomy, Collaborative Innovation Center for IFSA (CICIFSA), Shanghai Jiao Tong University, Shanghai 200240, China
e-mail: fhe@sjtu.edu.cn

CAS Center for Excellence in Ultra-intense Laser Science, Shanghai 201800, China

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1.1 Introduction

Modern light-matter interaction researches date back to Einstein's explanation of the photoelectric effect, in which ionization happens only if the absorbed photon energy is larger than the binding energy. The advent of laser technologies has boosted light-matter interaction researches into a new era, where novel nonperturbative phenomena are discovered, for example, strong-field tunneling ionization [1], above-threshold ionization [2], high-harmonic generation [3–5], nonsequential double ionization [6], low-energy structures [7, 8], and photoelectron holography [9]. Among these fascinating scenarios, stabilization of atoms in intense laser fields, i.e., the counterintuitive decrease of the ionization probability with the increase of driving laser intensities, attracts the attention of the ultrafast community [10–12]. Two mechanisms are known for ionization stabilization. One is interference stabilization [13, 14], in which the released electron wave packets from populated Rydberg states interfere destructively. The other is adiabatic stabilization, in which the multiphoton ionization is suppressed due to the deformation of Kramers-Henneberger (KH) wave functions [15–18], which are defined to be the eigenstates of a time-averaged Hamiltonian [19].

Though theoretically predicted for decades, the experimental confirmation of adiabatic stabilization is obscure due to ionization depletion and the focal-intensity average of lasers. In real experiments, the fine structure related to the stabilization may be smeared out after integrating all ionized fragments driven by different laser intensities. Furthermore, while the field strength in the focused center reaches the threshold of stabilization, the lower intensity around the focusing spot may completely ionize the target. The target might also be completely ionized before the laser field reaches its peak intensity in the time domain [20]. Up to now, there is only tantalizing indirect experimental evidence [21, 22] for the adiabatic stabilization. For example, in [23], a large acceleration of neutral atoms was reported and regarded as a signal of stabilization [18]. However, this evidence is not convincing enough as frustrated ionization [24], in which the ionized electrons get recaptured by the parent nuclei, has similar output. The ionization stabilization of Rydberg atoms [25] is not convincing evidence since the nonadiabatic coupling [26, 27] in intense fields populates a superposition of Rydberg states thus the ionization suppression might be attributed to the interference stabilization [13].

There are vast researches on adiabatic stabilization [10–12]. However, only a few attempted to directly identify KH states. Popov et al. [28] proposed to affirm the existence of the KH states via the energy shift of the photoelectron [29–31]. Morales et al. identified specific fine structures in photoelectron momentum distribution contributed by excited KH states [32]. Jiang et al. suggested that the photoelectron momentum distribution carrying dynamical interference structures provides information on adiabatic stabilization [33]. However, these proposals are sensitive either to the laser intensity or to the pulse envelope and are not robust against ionization depletion. Thus, the experimental realization is still challenging.

In one of our recent publications, we discussed the possibility of realizing Young's double-slit experiment with a single atom via KH states [34]. Here, we proposed to detect KH states using a bichromatic pump-probe strategy, in which the KH state is formed by the pump pulse and ionized by the probe one. By detecting the photoelectron momentum distribution, one is able to extract the dichotomic structure of the target, and thereby affirm the existence of KH states. The spin flipping for atoms following a loop in the laser parameter space provides an alternative route.

1.2 Models and Methods

1.2.1 Dipole Kramers-Henneberger Transformation

Our start point is the time-dependent Schrödinger equation (TDSE) (atomic units are used throughout unless stated otherwise)

$$i\frac{\partial}{\partial t}\psi(\mathbf{x},t) = H\psi(\mathbf{x},t), \tag{1.1}$$

where $H = \frac{1}{2}\left(\mathbf{p}+\mathbf{A}(t)\right)^2 + V(\mathbf{x})$ for a dipole laser field. The Kramers-Henneberger (KH) transformation [19] provides a comoving frame for a free electron interacting with laser pulse field, which is implemented via a time-dependent translation operator

$$U_d = \exp\left(i\mathbf{p}\cdot\boldsymbol{\beta}(t)\right)\exp\left(-i\int^t d\tau \mathbf{A}^2(\tau)/2\right). \tag{1.2}$$

Define $\psi_{KH} = U_d\psi$, the Hamiltonian transforms into the form

$$H_d^{KH} = \frac{1}{2}\mathbf{p}^2 + V(\mathbf{x}+\boldsymbol{\beta}(t)), \tag{1.3}$$

here $\boldsymbol{\beta}(t) = \int^t d\tau \mathbf{A}(\tau)$ is the displacement of the photoelectron. For a linearly monochromatic plane wave laser field with a frequency ω, the displacement is given by

$$\boldsymbol{\beta} = \boldsymbol{\beta}_0 \sin(\omega t), \tag{1.4}$$

with $\boldsymbol{\beta}_0 = \beta_0\mathbf{e}_x$ and $\beta_0 = \frac{E_0}{\omega^2}$. One can thus expand the potential into Fourier series

$$V(\mathbf{x}+\boldsymbol{\beta}(t)) = \sum V_n(\mathbf{x};\boldsymbol{\beta}_0)e^{-in\omega t}. \tag{1.5}$$

The harmonic component is given by $V_n(\mathbf{x}) = \int_0^{2\pi} d\phi V\left(\mathbf{x}+\boldsymbol{\beta}(\phi)\right)e^{in\phi}/(2\pi)$. Ionization induced by the nonzero component is suppressed with increasing ω [35]. These observations inspire the concept of KH atom, *i.e.*, the atom in a series of

nontrivial metastable states that exist only when in the laser field. KH atom is the eigenstate of the Hamiltonian

$$H_0^{KH} = \frac{1}{2}\mathbf{p}^2 + V_0(\mathbf{x}; \boldsymbol{\beta}_0) \tag{1.6}$$

and is closely related to the ionization stabilization, see Sect. 1.2.3 for more discussions.

1.2.2 Nondipole Kramers-Henneberger Transformation

Before going into the detailed discussion of the KH states, here we study the Kramers-Henneberger transformation in its general form first. Førre et al. [36] generalized the Kramers-Henneberger transformation to include the nondipole effect when the laser pulse is monochromatic. For a complete discussion of the nondipole effect in the nonrelativistic regime, we further consider the case when the laser vector potential is given by the superpositions of propagating waves

$$\mathbf{A}(t, \mathbf{x}) = \sum_a A^a(t_-^a), \tag{1.7}$$

where t_-^a is the light-front time

$$t_-^a = t - \mathbf{x} \cdot \mathbf{n}^a / c \tag{1.8}$$

of the a-th pulse and $\mathbf{n}^a$ is the pulse's propagating direction. We use axial gauge $\mathbf{n}^a \cdot \mathbf{A}^a = 0$. The field decomposition follows from $\mathbf{E} = -\partial_t \mathbf{A} = \sum_a \mathbf{E}^a(t^a)$ and $\mathbf{B} = \nabla \times \mathbf{A} = \sum_a \mathbf{B}^a(t^a)$.

As the time dependent displacement $\boldsymbol{\beta}(t, \mathbf{x}) = \sum_a \int_{-\infty}^{t^a} d\tau_- \mathbf{A}^a(\tau_-^a)$ is spatially dependent, we need to properly order the operator when defining the nonuniform KH transformation as

$$\begin{aligned} U &=: \exp\left(i\boldsymbol{\beta}(t, \mathbf{x}) \cdot \mathbf{p}\right) : \\ &= 1 + i\beta^i p_i + \frac{i^2}{2!}\beta^i\beta^j p_i p_j + \frac{i^3}{3!}\beta^i\beta^j\beta^k p_i p_j p_k \ldots, \end{aligned} \tag{1.9}$$

where the Einstein summation rule is adopted for repeated indices in (1.9).

The commutators between the KH transformation operator U and momentum and position operators are summarized as follows

$$\begin{aligned} [U, \mathbf{x}] &= \boldsymbol{\beta}(t, \mathbf{x}) U \\ \left[U, \mathrm{p}^\mu\right] &= \sum_a \frac{\mathrm{n}^{\mu a}}{c} (\mathfrak{A}^a \cdot \mathbf{p}) U \end{aligned} \tag{1.10}$$

here $\mathrm{n}^{\mu a} = (1, \mathbf{n}^a)$ and $\mathrm{p}^{\mu} = (i\frac{\partial}{c\partial t}, -i\frac{\partial}{\partial \mathbf{x}})$. $\mu = 0$ gives time component and $\mu = 1, 2, 3$ gives the spatial component. $\mathfrak{A}^a$ is defined via series,

$$\begin{aligned}\mathfrak{A}^a(t, \mathbf{x}) = \mathbf{A}^a(t_-^a) &+ \sum_b \mathbf{A}^a(t_-^a)\cdot\mathbf{n}^b\mathbf{A}^b(t_-^b)/c \\ &+ \sum_{b,d} \mathbf{A}^a(t_-^a)\cdot\mathbf{n}^b\mathbf{A}^b(t_-^b)\cdot\mathbf{n}^d\mathbf{A}^d(t_-^d)/c^2 \\ &+ \sum_{b,d,f} \mathbf{A}^a(t_-^a)\cdot\mathbf{n}^b\mathbf{A}^b(t_-^b)\cdot\mathbf{n}^d\mathbf{A}^d(t_-^d)\cdot\mathbf{n}^f\mathbf{A}^f(t_-^f)/c^3 + \cdots\end{aligned} \tag{1.11}$$

When all fields propagate collinearly, $\mathfrak{A}^a$ equals $\mathbf{A}^a$. Let $\psi_{KH} = U\psi$, the equation of motion for ψ_{KH} is given by

$$\begin{aligned}i\frac{\partial}{\partial t}\psi_{KH} &= H_{KH}\psi_{KH} \\ &= H\psi_{KH} + \left[i\frac{\partial}{\partial t} - H, U\right]U^{\dagger}\psi_{KH}.\end{aligned} \tag{1.12}$$

H_{KH} can be calculated by the commutating relations (1.10). Define

$$\mathbf{x}_{KH} = \mathbf{x} + \boldsymbol{\beta}(t, \mathbf{x}), \tag{1.13}$$

the transformed Hamiltonian is

$$\begin{aligned}H_{KH} \approx &\frac{1}{2}(\mathbf{p}^2 + \mathbf{A}(\mathbf{x}_{KH}, t)^2) + V(\mathbf{x}_{KH}) \\ &+ \sum_a \frac{1}{c}\left[(\mathbf{p} + \mathbf{A}(t))\cdot\mathbf{n}^a(\mathbf{A}^a(t)\cdot\mathbf{p}) + \boldsymbol{\beta}(t)\cdot\mathbf{n}^a\mathbf{E}^a(t)\cdot\mathbf{p}\right],\end{aligned} \tag{1.14}$$

here we expand H_{KH} to the order of 1/c,

When the laser field is monochromatic, (1.14) is identical to the results obtained in [36]. New interacting terms $\mathbf{A}(t)\cdot\mathbf{n}^a\mathbf{A}^a(t)\cdot\mathbf{p}$ and $\boldsymbol{\beta}(t)\cdot\mathbf{n}^a\mathbf{E}^a(t)\cdot\mathbf{p}$ appear when there are multi-color non-collinearly propagating lasers. The nice feature of H_{KH} in (1.14) is that there are no coupling between momentum operators and spatial-dependent functions, thus one can use a fast Fourier transformation (fft) based split-operator algorithm to solve the TDSE.

Unfortunately, (1.14) is not suitable for studying nondipole effects when $\boldsymbol{\beta}(t)\cdot\mathbf{n}^a\mathbf{E}^a(t)\cdot\mathbf{p} \neq 0$, as $\boldsymbol{\beta}(t)$ is not always a small quantity. However, this difficulty could be avoided in the electric field gauge [39].

With the gauge transformation $\psi^L = \exp(i\mathbf{x}\cdot\mathbf{A}(t, \mathbf{x}))\,\psi$, the Hamiltonian in the electric field gauge reads

$$H^L = \tfrac{1}{2}\mathbf{p}^2 + V_C(\mathbf{r}) + \mathbf{x}\cdot\mathbf{E} - \textstyle\sum_a \mathbf{x}\cdot\mathbf{E}^a\mathbf{p}\cdot\mathbf{n}^a/c \tag{1.15}$$

Define the time dependent displacement

$$U_{KH}^{L} = \exp\left(\sum_{a} \mathbf{x}\cdot\mathbf{A}^{a}(t)\mathbf{p}\cdot\mathbf{n}^{a}/c\right), \tag{1.16}$$

and

$$\psi_{KH}^{L} = U_{KH}^{L}\psi^{L}, \tag{1.17}$$

we have the transformed Hamiltonian

$$\begin{aligned} H^{L} = {} & \frac{1}{2}\mathbf{p}^{2} + V_{C}\left(\mathbf{r} + 1/c\sum_{a}\mathbf{n}^{a}(\mathbf{x}\cdot\mathbf{A}^{a}(t))\right) + \mathbf{x}\cdot\mathbf{E}(t,\mathbf{x}) \\ & + \frac{1}{c}\sum_{a}\left[(\mathbf{E}(t)\cdot\mathbf{n}^{a})(\mathbf{x}\cdot\mathbf{A}^{a}(t)) - (\mathbf{p}\cdot\mathbf{n}^{a})(\mathbf{p}\cdot\mathbf{A}^{a}(t))\right]. \end{aligned} \tag{1.18}$$

1.2.3 The Kramers-Henneberger States

The KH states are defined to be the eigenstates of the KH Hamiltonian. Generally speaking, they are metastable states in the laser field while their stability increase when the laser frequency increases [35]. Thus, we expect the KH states could play an important role in the high frequency laser field. The dynamical information manifests itself already in (1.5). The adiabatic potential V_0 gets deformed by the laser field, which in turn deforms the bound state, and the nonzero order harmonic terms ionize the KH states. As we will see in Sect. 1.2.4, the above picture is extremely useful when the laser frequencies are high.

If the laser pulse is linearly polarized, V_0 has a dichotomic structure [17, 40]. Figure 1.1a plots V_0 when $\beta_0 = 10$ a.u., from which we see two local minimum located at $\pm\boldsymbol{\beta}_0$. Compared with the laser free case, V_0 is no longer isotropic and has only axial symmetries. As a consequence, the orbital angular momentum number is not conserved and the KH states could be labeled in the same manner as the homonuclear diatomic molecules. Due to the spin-orbital coupling, only the total magnetic moment is conserved. We will discuss the role of the spin-orbital coupling in Sect. 1.3.4 when we deal with the geometric phase.

The potential V_0 depends on the parameter $\boldsymbol{\beta}_0$, which means the KH states and their eigenenergies depend also on $\boldsymbol{\beta}_0$. We plot the eigenenergies of the ground state KH states in Fig. 1.1b. The eigenenergies increase when β_0 increases [29–31].

The adiabatic potential V_0 is dichotomic, so is the wave packet [16], see Fig. 1.1c. The wave function is localized at $\pm\boldsymbol{\beta}_0$ and has similar properties as homonuclear diatomic molecules. Thus, we could have charge resonance enhanced ionization [34, 37, 38] in atoms.

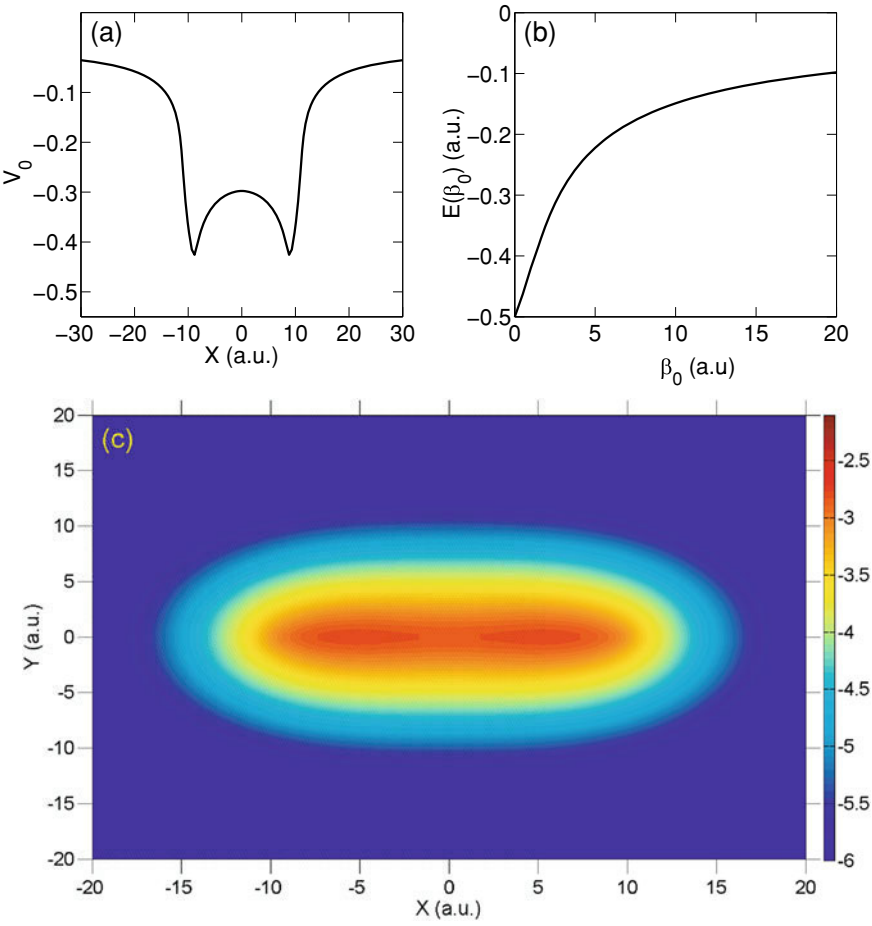

Fig. 1.1 The laser field is linearly polarized along the x-axis. **a** The plot of V_0 when $\beta_0 = 10$ a.u. **b** The eigenenergies of the ground state KH hydrogen atoms as a function of β_0. **c** The probability distribution of the ground state KH atom when $\beta_0 = 10$ a.u.

1.2.4 Dynamics of Kramers-Hennerberger States

In practice, we need to consider the effect of the pulse envelope. Thus, the expression for the displacement in (1.4) is replaced by

$$\boldsymbol{\beta}_0 = \beta_0 f(t)\mathbf{e}_x. \tag{1.19}$$

The corresponding laser field is given by $\mathbf{E}(t) = -\frac{\partial^2}{\partial t^2}\boldsymbol{\beta}$. We use the envelope $f(t) = \cos^2(\pi t/L)$ $(-L/2 < t < L/2)$ throughout this paper, where L stands for the pulse duration. The ground state of (1.1) is obtained using the imaginary time method [41],