Elements of Photoionization Quantum Dynamics Methods

Elements of Photoionization Quantum Dynamics Methods

Lampros A A Nikolopoulos
School of Physical Sciences, Dublin City University, Dublin, Ireland

Morgan & Claypool Publishers

ISBN 978-1-68174-712-5 (ebook)
ISBN 978-1-68174-713-2 (print)
ISBN 978-1-68174-714-9 (mobi)

DOI 10.1088/2053-2571/aaf10d

Version: 20190301

IOP Concise Physics
ISSN 2053-2571 (online)
ISSN 2054-7307 (print)

A Morgan & Claypool publication as part of IOP Concise Physics
Published by Morgan & Claypool Publishers, 1210 Fifth Avenue, Suite 250, San Rafael, CA, 94901, USA

IOP Publishing, Temple Circus, Temple Way, Bristol BS1 6HG, UK

This book is dedicated to Natasa and my parents, Thano and Andriana.

Contents

Prologue

The first principle is that you must not fool yourself—and you are the easiest person to fool. So you have to be very careful about that. After you've not fooled yourself, it's easy not to fool other scientists. You just have to be honest in a conventional way after that.

Richard Feynman, Cargo-Cult Science, Caltech 1974

My intention in writing this book was to revisit some theoretical methods related to the interaction of electro-magnetic fields with elementary quantum systems. From a broader viewpoint, this scientific area is perhaps the oldest one (in fact, the black-body radiation problem boosted the birth of quantum mechanics as a physical theory of atomic-sized systems) and as such it is a very mature field. So, in principle, there are not many topics left to talk about without actually revisiting them. After ninety years of quantum mechanics and about sixty years from the firing-up of the first laser, most theoretical methods of relevance are probably already known. Now and then some of them re-appear under different names, but a closer look reveals that it is not in fact a question of a 'first-seen' approach. Thus, why take on the hassle and write another book on a not-so-modern subject (and consequently not an interesting topic among the leaders of technology and fund hunters)? One answer is that I have the feeling that it is precisely this wealth of, seemingly different, methods and approaches that constitutes a serious obstacle for newcomers to this field (and others). The connection of the (normally) heavily formalized methods with the simpler physical principles is fuzzy and almost never mentioned in the specialized literature, as it is taken for granted, or, when it is referenced in the form of a citation, it soon leads to a stochastic branching process problem in tracing the labyrinthine literature. As a result, although quick access to the scientific literature is now guaranteed, in my view this is often not helpful in clarifying matters and eventually the underlying physical context is left in the distant past. This why the phraseology of the research literature sometimes looks like the reinvention of the wheel. In a way, this is due to the re-ignited interest in practical calculations following the development of lasers with unprecedented features, such as durations as short as a few attoseconds (10^{-18} s), with UV and soft x-ray wavelengths (free-electron lasers), and extremely high brilliance. Thus, the topic of this book may not yet be an outdated subject. A second reason for writing this book is more practical, as I am not lucky enough to teach an undergraduate quantum mechanics course, most of the (re-occurring) questions I have faced are related to the problem of how to introduce my MSc and PhD students to the subject. This is a persistent problem as it takes time to develop and maintain such expertise (my experience at various institutions points to varying approaches). For example, questions such as: can a quantum system *emit* a photon and increase its energy? Does this need to be taken into account? How is the classical (wave) description of the field justified when its interaction takes place in terms of photon absorptions/emissions? How is it possible for electrons to absorb photons even if they are 'free', or are the photons localized? To make a long story short, this book is intended for newcomers who would like to have a introductory (soft) crash-course in ultra-short laser–atomic dynamics methods. The computational implementations of all of these methods are discussed in detail in the literature (with the exception of the inherently (highly) non-linear neural networks methods) and only an incomplete account will be provided here. Good knowledge of quantum mechanics is necessary. Taking as a measure the majority of physics courses worldwide, it is expected that the reader will have some familiarity with time-dependent treatments, continuum (scattering) states, and feel comfortable with differential calculus (including the Fourier transformation and the Dirac-delta function) and basic probability theory concepts such as mean value, standard deviation and covariance (or correlation).

Unfortunately, due to the necessity of brevity, not all of the subjects I would like to discuss are included; many are left out or relegated to the supplementary chapters 11 and 12, or to a future expanded version (if the opportunity should arise). Some of these omitted subjects include: elliptically polarized lasers; the Dirac treatment of hydrogen; the (relativistic) multipole expansion of the laser–atom interaction; the derivation of the photoionization boundary condition from a time-dependent scattering description; a more complete presentation of density matrix theory; a basic exposition of the quantum theory of light and its interaction with atomic systems; a more systematic presentation of ionization with randomly fluctuating fields; and a more sophisticated treatment of discrete (box-normalized) states as localized wavepackets. Also, some detailed applications for atomic hydrogen, molecular hydrogen ions, quantum dots and molecular dissociation have also been left to future updates of the supplementary chapters. Possibly, the most important omissions are a decent presentation of the three-step tunneling ionization model for long-wavelength lasers and an *ab initio* theory of the interaction of lasers with two or more electron atomic systems. Consistent with the book's title, attention is mainly given in the theoretical methods to tackling quantum dynamics, rather than to the description of atomic dynamics itself. Among the methods presented, the simplest ones are discussed, but not all to the same degree of sophistication. Limitations of space have shortened and compacted some discussions, although more clarity was possibly needed. In these cases the presentation relies on some extra mental effort from the reader. Usually one has to make a choice as to how to present a subject as several viewpoints may be available; here my primary criterion was to choose the most elementary possible (while retaining accuracy in the description) as well as what appeared to most logically emerge from quantum mechanics principles. Usually, the material included in the earlier chapters will be used in later chapters, but in a very few cases I have included some additional discussion for reasons of completeness.

The current status of the research field dealing with ultra-short laser–matter dynamics is state-of-the-art in terms of theoretical and numerical methods, with all the elements of excellence and elegance, but it was not appropriate to use the original scientific literature, as a fair reference treatment was impossible. I have tried to include citations to textbooks, in particular the classic ones, with minimal reference to more modern (and specialized) textbooks. Again, for the interested reader who wants to have a starting point for past and (most) modern theoretical activities in the fields of strong and ultra-short laser fields, I can mention (in approximate alphabetical order) those theoretical researchers whom I have personally advised in past years; these include the works on *ab initio* theories of atoms and molecules in strong-fields by L Argenti, H Bachau, J Burgdörfer, P Decleva, J Feist, P Lambropoulos, E Lindroth, F Martin, L Madsen, A Palacios, B Piraux, K Taylor, H Van der Hart and A Saenz; the works by A N Grum-Grzhimailo, N Rohringer and R Santra on free-electron laser x-ray ionization processes; and finally the works by M Ivanov and C D Lin on tunneling ionization processes.

How to approach this book

Now, I would like to comment from a more general standpoint on how the reader should approach studying this book. In the vast majority of cases, lectures, textbooks or other means of presenting a subject inevitably include a carefully polished outcome of the (usually) collective and intensive research efforts of people who have followed different routes to tackle the subject. This is the standard way in which fundamental research proceeds, so what we see are actually the boulevards and not the painful dead-ends. It is only when one studies a subject thoroughly that one is really armed to explore the by-roads and even to explore one's own, lonely and harsh, paths—most of them ending wastefully at nowhere. As no royal roads to learning exist, a simple joyful read (such as a historical novel) will never make you an expert; quite the opposite, it only leads to fooling yourself and harms your scientific integrity. *Thorough* study must always be combined with a persistent, explorative (researching) open-minded spirit. Being open to other views and ideas—this is what is defined as an open mind—but do it truly, not only in theory. Maybe all these are already well understood and known, but if you happen to read what is in this book, do not be satisfied with the easier route; instead of simple reading my words, make it your goal to practice and check everything. In any objections you may have, it is advisable to use science as accurately and clearly as you can, because it is on your own two feet that you need to stand respectfully.

Acknowledgments

As writing this book was an entirely home-based activity, it goes without saying that I have enjoyed writing against the noisy background of my children, Andriana, Thanos and Foteini. It was a slow-paced process, not free from periodical update requests from the gentle Jeanine Burke. Last but not least, it is fair to mention that I (and indirectly this book) have undoubtedly benefited in different ways (along with their wit) from my past and current research colleagues.

Dublin, November 2018

Author biography

Lampros Nikolopoulos

Lampros Nikolopoulos, PhD, is a lecturer at the School of Physical Sciences at Dublin City University (DCU). He was brought up in Greece and holds a BSc (Hons) in physics from the Physics Department of the University of Athens, and an MSc and PhD in theoretical atomic physics from the University of Crete, Greece. His previous posts include MPQ-Garching in Germany, IFA-Aarhus in Denmark, and QUB-Belfast in the UK, before he settled in Dublin. His research interests include ultra-short laser–matter quantum dynamics and development of high-performance computational methods. A recent thesis supervised by him received a prize from the UK-IOP Computational Group as the 'best PhD thesis in Computational Physics' for the year 2016. He has (co)authored over 80 journal articles, two book chapters and co-edited a special issue on 'Short-wavelength free-electron lasers'.

Glossary of symbols

Acronym	Meaning
AC	autocorrelation
AIS	autoionization state
ATI	above-threshold ionization
a.u.	atomic units
BC	boundary condition
BCH	Baker–Campbell–Hausdorff
CI	Configuration interaction
CSOP	complete set of physical operators
DM	density matrix
DME	density-matrix equation
EM	electro-magnetic
EMF	electro-magnetic field
FEL	free-electron laser
FT	Fourier transform
GVD	Group velocity dispersion
HHG	high-harmonic generation
HO	harmonic oscillator
IP	interaction picture
laser	light amplification by stimulated emission radiation
l.h.s.	left-hand side
ME	Maxwell's equation
ODE	ordinary differential equation
OU	Ornstein–Uhlenbeck
PAD	photo-electron angular distribution
PES	photo-electron kinetic spectrum
QDE	QM dynamics equation
QM	quantum mechanics
RWA	rotating-wave approximation
r.h.s.	right-hand side
SE	Schrödinger equation
SH	spherical harmonics
SP	Schrödinger picture
SVA	Slowly varying amplitude
TD	time-dependent
TDDM	time-dependent density matrix
TDSE	time-dependent Schrödinger equation
TLS	two-level system
WKB	Wentzel–Kramers–Brillouin

Symbol	Description
$\hat{H}_0$, $\hat{H}_a$	atomic Hamiltonian operator
$\hat{V}_a$	atomic (molecular) potential
$\psi(\mathbf{r}, t)$	time-dependent wavefunction
$\phi(\mathbf{r})$	time-independent wavefunction

$\mathbf{E}(t)$	semiclassical electric field after dipole approximation
$\mathcal{E}(t)$, $\mathcal{E}_0$	electric field envelope, amplitude
$\hat{D}(t)$	dipole interaction operator
$\delta(\omega)$, $\delta_T(\omega)$	Dirac-delta function, finite time Dirac-delta
$\rho(\mathbf{r}, t)$	density-matrix operator
ω_L, ω	laser-field central carrier frequency
ω_0	atomic energy variable, two-level energy difference, HO's energy
c	light speed in a vacuum
$\mathcal{I}(t)$	pulse's envelope intensity $\mathcal{I}(t) = \lvert\mathcal{E}(t)\rvert^2$
$\mathcal{C}_i(t, t')$	first ($i = 1$) and second ($i = 2$) autocorrelation (coherence) functions of $\mathcal{E}$