AFRL-AFOSR-JP-TR-2021-0008

Quantum collision theory for photon scattering from atoms and molecules

Igor Bray
CURTIN UNIVERSITY OF TECHNOLOGY
BLDG 100, KENT STREET
BENTLEY, ,
AU

08/02/2021
Final Technical Report

Air Force Research Laboratory
Air Force Office of Scientific Research
Asian Office of Aerospace Research and Development
Unit 45002, APO AP 96338-5002

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*	**2. REPORT TYPE**	**3. DATES COVERED** *(From - To)*
02-08-2021	Final	10 May 2019 - 09 May 2021

4. TITLE AND SUBTITLE
Quantum collision theory for photon scattering from atoms and molecules

5a. CONTRACT NUMBER
FA2386-19-1-4044

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
Igor Bray

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
CURTIN UNIVERSITY OF TECHNOLOGY
BLDG 100, KENT STREET
BENTLEY,
AU

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AOARD
UNIT 45002
APO AP 96338-5002

10. SPONSOR/MONITOR'S ACRONYM(S)
AFRL/AFOSR IOA

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AFRL-AFOSR-JP-TR-2021-0008

12. DISTRIBUTION/AVAILABILITY STATEMENT
A Distribution Unlimited: PB Public Release

13. SUPPLEMENTARY NOTES

14. ABSTRACT
See attached

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**	**19a. NAME OF RESPONSIBLE PERSON**
a. REPORT	**b. ABSTRACT**	**c. THIS PAGE**			CHRISTOPHER VERGIEN
U	U	U	SAR	4	**19b. TELEPHONE NUMBER** *(Include area code)* 315-227-7002

Final Report for AOARD Grant FA2386-19-1-4044

"**Quantum collision theory for photon scattering**"

14 July 2021

Name of Principal Investigators (PI and Co-PIs): Prof. Dmitry Fursa, Prof. Igor Bray

- e-mail address : d.fursa@curtin.edu.au
- Institution : Curtin University
- Mailing Address : Department of Physics and Astronomy, Curtin University, GPO Box U1987, Perth, Western Australia, 6845
- Phone : +61 8 92662377
- Fax : +61 8 92662377

Period of Performance: 05/10/2019 – 05/09/2021

Abstract: Photon scattering and photoionization are fundamental processes that are central in modeling physical and technological problems occurring in spectroscopy, imaging, and communications. Starting from the fundamental principles of quantum electrodynamics we have developed a novel computational technique to study Rayleigh and Raman scattering and photoionization. This technique has been applied to study photon-induced processes in alkali atoms (Li, Na, K, Rb, Cs). A comprehensive set of cross sections has been determined for Rayleigh, Raman, and photoionization processes involving the ground and a large number of excited states of the atoms.

Introduction: Propagation of light in media (e.g. atmosphere) is an important physical problem with applications to spectroscopy, imaging, and communications. Modeling of light interaction with media requires an understanding of light scattering on, and excitations off, constituent atoms and molecules. The most consistent and accurate approach to model these processes is based on the application of quantum electrodynamics (QED). The development of the Kramers-Heisenberg-Waller (KHW) matrix elements provided a clear description of photon scattering processes up to second order in perturbation theory. The difficulty in calculating the KHW matrix elements is related to an account of the continuum spectrum of the target where the pole terms have to be dealt with. Most accurate calculations are restricted to the case where the Green's function may be given analytically which limits those techniques to hydrogen-like systems. The full QED analysis of light scattering from complex atoms and molecules is significantly lacking.
The major aim of this project is to develop a practical and computationally reliable technique for the evaluation of the KHW matrix elements that in turn allows performing a comprehensive study of photon collision processes with atoms and molecules. This technique is to be tested against accurate results that are available for a relatively simple system, such as hydrogen atom and alkali atoms. The goal is to extend this technique to photon processes involving more complex and important in applications systems: molecular hydrogen and hydrogen storage, quantum bits using Rb atoms and quantum computing, entangled photon scattering and quantum illumination and radar, etc.

Method: In all suggested implementations of quantum radar, and applications in imaging, spectroscopy, communications, radiation transport, and opacity the underlying physics is based on photon interactions with media, that is with atoms and molecules that make the media. Such interactions are governed by the laws of quantum electrodynamics. QED is the relativistic theory that describes the dynamics of photons and their interaction with matter. Accurate description and modeling of these processes have to be done within the laws of QED and represent a formidable challenge. Within QED the photon-atom scattering can be described by the well-known Kramers-Heisenberg-Waller (KHW) second-order amplitude [17,18] for the transition from an initial state |i> to a final state |f>

$$M_{fi} = \epsilon \cdot \epsilon'^{*} \langle f | e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}} | i \rangle - \sum\!\!\!\!\!\!\int_{t} \left[\frac{\langle f | e^{-i\mathbf{k}'\cdot\mathbf{r}} (\epsilon'^{*} \cdot \mathbf{p}) | t \rangle \langle t | e^{i\mathbf{k}\cdot\mathbf{r}} (\epsilon \cdot \mathbf{p}) | i \rangle}{E_t - E_i - \omega - i0} + \frac{\langle f | e^{i\mathbf{k}\cdot\mathbf{r}} (\epsilon \cdot \mathbf{p}) | t \rangle \langle t | e^{-i\mathbf{k}'\cdot\mathbf{r}} (\epsilon'^{*} \cdot \mathbf{p}) | i \rangle}{E_t - E_i + \omega'} \right],$$

where the summation is over all intermediate (bound and continuum) states |t>.

The main difficulty in photon scattering from atoms and molecules is the same as with electron collisions: how to account for the infinite number of various reaction channels (summation over the intermediate states |t> in the KHW amplitude). The technique to address this problem we have developed for electron collisions is equally applicable for photon scattering. Namely, we need to represent the whole spectrum of the target by a finite size expansion using a Sturmian basis.
We have developed two techniques to model photon collisions with quasi one-electron atoms [1,2]. One method (PV) is based on direct integration over the continuum spectrum of the target and another method is based on an application of the Complex Exterior Scaling technique (CES). Alkali atoms (Li,…, Cs) are well described by a model of one active electron above a frozen core and allow both PV and CES techniques to be applied to study photon scattering processes. For heavy atoms, Rb and Cs, relativistic effects are important. To model accurately such systems we have extended the PV method to a fully relativistic domain [3] Dirac L-spinors as Sturmian functions and formulating the problems starting with Dirac Hamiltonian. To account for the effects of frozen-core electrons more accurately we have used model polarization potentials and a modified form of the dipole operator [2,3].

Results and Discussion:

Comprehensive results have been obtained for Rayleigh and Raman scattering and photoionization cross sections for H, Li, Na, K, Rb, Cs atoms. We demonstrated the importance of relativistic effects for heavy atoms and the importance of the account of core polarization. In particular, for photoionization processes, we have shown that the Cooper minimum position is strongly affected by the choice of the modified dipole operator. We have compared our results with accurate photoionization cross sections of Zatsarinny and Tayal [Phys. Rev. A 2010, 81, 043423] produced using a Dirac-based B-spline R-matrix method which uses the most sophisticated atomic structure model that has been used for single-photon ionization of an alkali atom. We have shown that our photoionization cross sections are in good agreement with the results of Zatsarinny and Tayal if in our technique the polarization potentials and the modified dipole operator is chosen to fit the resonance transition of the atom (for example, for Cs atom this is the 6s – 6p transition). An illustration of the calculated Rayleigh and Raman scattering cross section for the Cs atom is presented in Fig. 1, while in Fig. 2 an example of photoionization cross sections is presented for the K atom.

The amount of collision data obtained during the project is very large. We are planning to submit a paper to Atomic Data and Nuclear Data Tables – this is a journal dedicated to providing access to such large datasets. We are also considering to produce a database for the calculated collision data – accessible free by any researcher. An example of such a database can be seen at mccc-db.org, where the results of electron-molecule scattering obtained in our group are presented.

The work done on this project over the last two years allowed us to progress sufficiently to offer a Ph.D. project on photon scattering. This Ph.D. project is taken by Adam Singor, who is the first author of the latest two papers published as the result of the project [2,3].
An exciting opportunity is to work on Raman spectroscopy of molecular hydrogen. There are two types of molecular hydrogen: ortho and para hydrogen. They have different specific heat and this is a crucial issue for hydrogen storage projects. As a quantum system, para and ortho hydrogen have different rotational spectrums that allow using Raman spectroscopy to measure their relative densities. The technology we have developed as a result of USAF grants: this current grant and previous grants on modeling electron collisions with molecular hydrogen will allow us to conduct an investigation in

Raman spectroscopy relevant for hydrogen storage. Here in Perth, there are several experimental and industry groups we will collaborate: Prof. Eric May is the group leader at the University of Western Australia and Managing Director, Future Energy Exports CRC Pty Ltd, Prof. Craig Buckley from Curtin University is the manager of the Hydrogen Storage Research Group (HSRG).
We have a long-standing collaboratin with researcger from Los Alamos National Laboratory (Mark Zammit, James Colgan, Chris Fontes). This project allowed us to extend it to photon-driven processes.

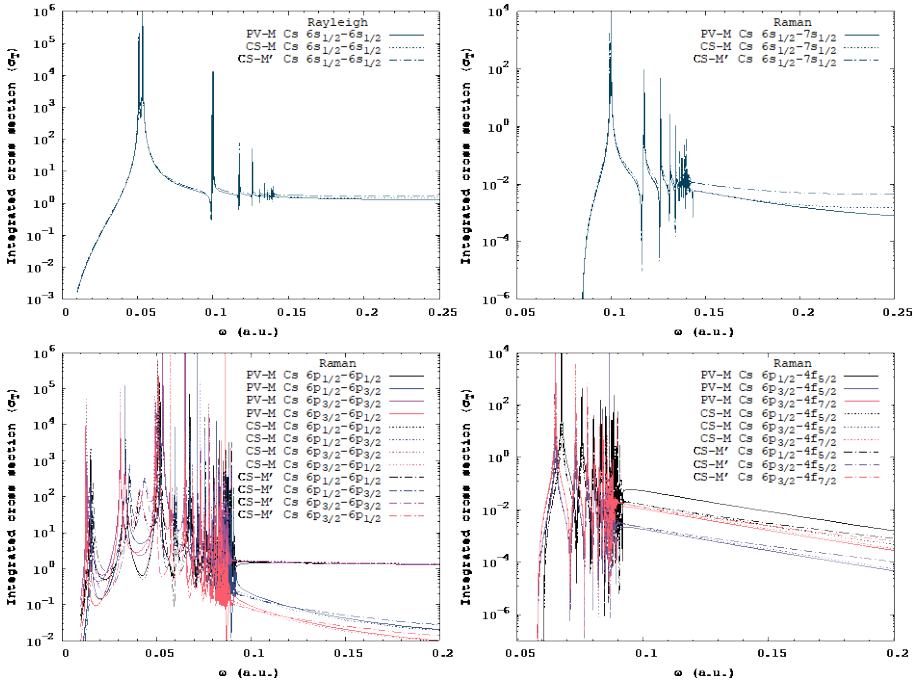

Figure 1. Rayleigh and Raman cross sections for scattering on the ground and first excited p state of cesium, see Ref [3] for more details.

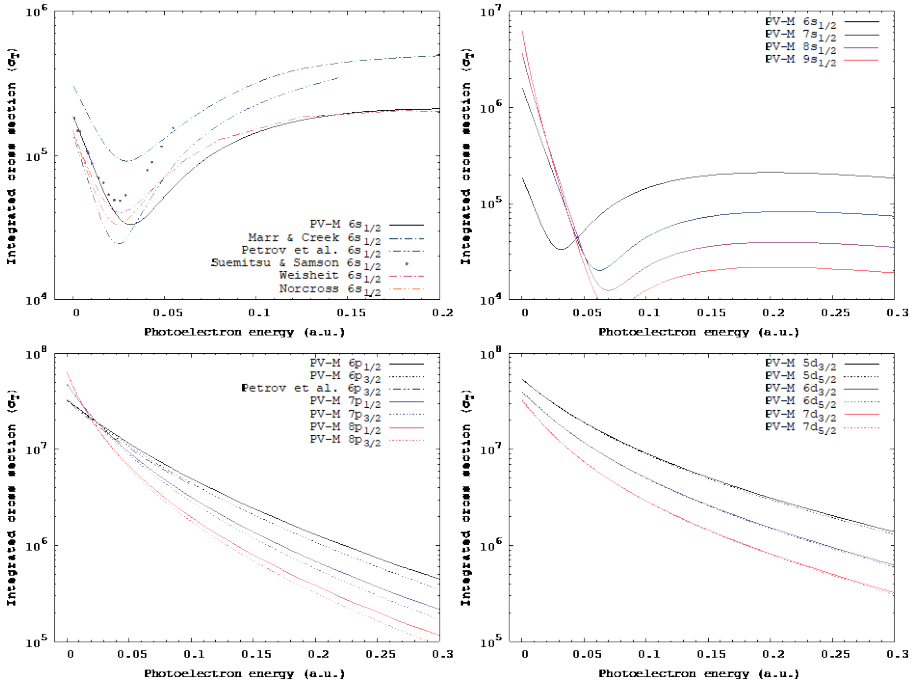

Figure 2. Photoionization cross sections for the ground and excited states of potassium, see Ref [3] for more details.

List of Publications and Significant Collaborations that resulted from your AOARD supported project:

a) papers published in peer-reviewed journals:

1. McNamara, K.; Fursa, D.V.; Bray, I. "Efficient calculation of Rayleigh and Raman scattering." Phys. Rev. A 2018, v. **98**, p. 043435.

2. Singor, A.; Fursa, D.; McNamara, K.; Bray, I. "Rayleigh and Raman Scattering from Alkali Atoms." Atoms 2020, v. **8**, p. 57.

3. Singor, A.; Fursa, D.; Bray, I. "A Fully Relativistic Approach to Photon Scattering and Photoionization of Alkali Atoms." Atoms 2021, v. **9**, p. 42.

d) conference presentations without papers:

1. International conference on photonic, electronic and atomic collisions (ICPEAC) 2019 – "Efficient Calculation of Rayleigh and Raman Scattering", K. McNamara, A. Singor, D. V. Fursa, I. Bray

2. Gaseous electronic conference (GEC) 2020 "Photon scattering from alkali atoms: Rayleigh, Raman, and photoionization processes", A. Singor, D. V. Fursa, K. McNamara, I. Bray
[This work was recognized with the best undergraduate poster award (attached).]

3. International conference on photonic, electronic and atomic collisions (ICPEAC) 2021 "A fully relativistic approach to photoionization of alkali atoms", A. Singor, D. V. Fursa, I. Bray

4. Gaseous electronic conference (GEC) 2021(upcoming, October) "A fully relativistic approach to photon scattering and photoionization for the alkali atoms", A. Singor, D. V. Fursa, R. P. McEachran, I. Bray

e) manuscripts submitted but not yet published, and

f) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.

D. V. Fursa, at the Lawrence Livermore National Lab, November 2019

Attachments: Publications a)

DD882: As a separate document, please complete and sign the inventions disclosure form.