ALI RAMADHAN

MMMGRUBS: MOLECULAR MOTION MOVIES AND GEOMETRY RECONSTRUCTION USING BAYESIAN STATISTICS

MMMGRUBS: MOLECULAR MOTION MOVIES AND GEOMETRY RECONSTRUCTION USING BAYESIAN STATISTICS

ALI RAMADHAN

Hakuna matata?

December 19, 2016

TECHNICAL ABSTRACT

Ever since the early days of Coulomb explosion imaging, the direct imaging of the structure and dynamics of individual molecules has been promised; however, in practice no one has been able to accurately retrieve this structure and all imaging relies on plotting the momentum vectors of the fragments in different ways to infer crude changes in geometry. The momentum vectors tell a large part of the story but do not provide a clear picture of the molecular dynamics everyone seeks so an accurate method of retrieving the structure is highly desirable. I use Bayesian inference and Markov chain Monte Carlo methods to elucidate the molecular geometries, allowing for the study of larger systems than before and for the inclusion of measurement error in these studies for the first time. Using this method, the ultrafast isomerization of carbonyl sulfide and acetylene is imaged in position-space and showcased as a molecular movie. The method, however, is much larger in scope and is generalized to any problem involving a physical system described by a system of ordinary differential equations, where the final conditions are experimentally measured and inference of the initial conditions is desirable.

TECHNICAL ABSTRACT

Insert better abstract.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

LISTINGS

ACRONYMS

Part I

HOW TO EXPLODE A MOLECULE

A lot goes into exploding a molecule, even the tiny ones featured here. To begin, a short laser pulse must be created and its interaction with the molecule must be understood. Once we have a short laser pulse and understand how it interacts, we can do Coulomb explosion imaging.

1 HOW TO MAKE A SHORT LASER PULSE

1.1 SPONTANEOUS AND STIMULATED EMISSION

1.2 PROPERTIES OF A LASER

1.3 PRODUCING LASER LIGHT

1.4 MAKING LASER PULSES

1.5 MAKING SHORT LASER PULSES

1.6 SHORT PULSE AMPLIFICATION

1.6.1 *Chirped pulse amplification*

1.6.2 *Optical parametric amplification*

1.7 LASER AMPLIFIERS

2

WHAT DOES A SHORT PULSE DO?

2.1 Short laser pulse properties

2.2 Interaction with single atoms

2.2.1 *Multiphoton ionization*

2.2.2 *Tunneling ionization*

2.2.3 *Keldysh parameter*

2.3 Interaction with single electrons

2.3.1 *Ponderomotive force*

2.3.2 *Relativistic Thomson scattering*

2.3.3 *Nonlinear Compton scattering*

2.4 Diatomic molecules in intense laser fields

2.5 Small polyatomic molecules in intelse laser fields

3 PUMP-PROBE COULOMB EXPLOSION IMAGING

3.1 EXPERIMENTAL SCHEME

In pump-probe Coulomb explosion imaging (CEI) one ultrashort laser pulse is split into two pulses through the use an asymmetric beam-splitter. One of the pulses, the pump pulse, is usually much weaker than the other, the probe pulse. A time delay τ between the pulses is then created such that the pump pulse goes first and the probe pulse second. The job of the pump pulse will be to initiate some change in the molecule. One example could include an isomerization of the molecule. Thus the pump pulse "pumps" the molecule into some excited state. The job of the powerful probe pulse is to engulf the molecule in an intense enough laser field such that multiple electrons are stripped off of it. The molecule's individual atoms are left in a highly-charged state and begin to behave as individual point charges in a purely Coulombic potential. The entire process occurs in the presence of a constant electric field and so the positively-charged ions all accelerate upwards towards a time- and position-sensitive detector. Thus the probe pulse allows for the "probing" of the excited state.

3.2 EXPERIMENTAL DETAILS

3.3 HISTORY AND ACCOMPLISHMENTS

CEI was first performed by Vager, Naaman, and Kanter (1989) whereby the Coulomb explosion was initiated by passing the molecule through a thin carbon film at high velocities.

3.4 OTHER WAYS OF INITIATING COULOMB EXPLOSIONS

Vager, Z., Naaman, R., and Kanter, E. P. (1989). Coulomb explosion imaging of small molecules. *Science* **244**, 426–431. This is a classic of linguistics.