

Announcing the Final Examination of Coleman Cariker for the degree of Doctor of Philosophy in Physics

Date: July 5, 2022

Time: 1:00 p.m.

Room: PSB 160

Dissertation title: Attosecond optical probe and control of atomic autoionizing states

Abstract:

Attosecond spectroscopy proves a particularly useful tool to monitor and control, in real time, the evolution of transiently bound electronic states in the atomic and molecular continuum. These transiently bound states, also known as autoionizing states, play a fundamental role in ionization and charge transfer processes in matter. This thesis is a theoretical study on the role of such autoionizing states in the ionization of polyelectronic atoms by sequences of ultrashort pulses, with particular reference to all-optical observables. To investigate various aspects of these new spectroscopies, we have adopted several complementary approaches. One approach is to solve the time-dependent Schrodinger equation (TDSE) numerically. We demonstrate a novel procedure for performing *ab initio* calculations, using an "essential states" basis, which is able to produce results identical to the full basis while decreasing the cost of calculation by as much as three orders of magnitude. We highlight the crucial role played by radiation-matter entanglement in multi-photon ionization processes, which has gone unrecognized so far. To do so, we extend the Jaynes-Cummings model, which concerns two isolated bound states in interaction with a monochromatic intense light pulse, to autoionizing states and demonstrate how quantum interference between autoionization and radiative ionization channels can be leveraged to control the lifetime of laser-dressed metastable states. The findings of the model are validated by *ab initio* calculations of transient absorption in argon, they are in excellent agreement with experimental measurements, providing support for the first observational evidence of a stabilization phenomenon predicted theoretically four decades ago. Recently, the powerful technique of four-wave mixing (4WM) has been extended to the XUV range, thus opening the way to background-free optical interferometry. We have devised the formalism needed to extrapolate, from the single-atom response, the off-axis dipolar emission from an extended sample, and implemented it to theoretically reproduce, for the first time and with semi-quantitative accuracy, four-wave mixing experimental spectra in helium and argon. Our essential-state methodology allow us to selectively remove individual resonances from a simulation. Using the 4WM software in conjunction with the essential-state propagator, therefore, we confirm how a non-collinear 4WM setup can measure the lifetime of dark transiently bound states.

Outline of Studies:

Major: Physics

Educational Career:

M. S. UCF 2015

B. S. UCF 2014

Committee in Charge:

Dr. Luca Argenti (Chair)

Dr. Michael Chini

Dr. Zenghu Chang

Dr. Konstantin Vodopyanov (External Committee Member)

Approved for distribution by Dr. Luca Argenti, Committee Chair, on June 14, 2022.

The public is welcome to attend.