Cross Border Workshop On Laser Science, 2006

University of Connecticut
Storrs, CT

Thursday, June 1

3:00 Opening remarks
George Gibson, University of Connecticut

Ultrafast biology – BSP 130

3:10 "The initial electron transfer reaction of photosynthesis: a new model for an old problem,"
Neal Woodbury, University of Arizona

3:55 "Direct observation of ultrafast dynamics in DNA bases,"
Helmut Satzger, NRC, Ottawa

4:25 "Detecting the dark states of xanthophylls using time-resolved absorption spectroscopy,"
Dariusz Niedzwiedzki, University of Connecticut

4:45 Break – lobby

Coherent control of biological systems – BSP 130

5:00 "Femtosecond coherent control for nonlinear spectroscopy and microscopy,"
Nirit Dudovich, NRC, Ottawa

5:30 "Ultrafast coherent spectroscopy of biological systems,"
Svetlana Malinovskaya, University of Michigan

6:15 Dinner – North West Dining Hall

7:30 Posters – lobby

Friday, June 2

Quantum control to wavefunction imaging – PB 120

9:00 "Measuring and controlling the birth of attosecond pulses,"
Nirit Dudovich, NRC, Ottawa

9:30 "Femtosecond multidimensional imaging - watching chemistry from the molecule's point of view,"
Oliver Gessner, NRC, Ottawa

10:00 Break – lobby

Entanglement and quantum imaging – BSP 130

10:15 "Dissociative wave packets in large molecules: control and measurement,"
Brett Pearson, Stony Brook

10:45 "Shaping potential energy surfaces with the dynamic stark shift,"
Ben Sussman, NRC, Ottawa

11:15 "Quantum tomography of cold atomic systems,"
Matt Partlow, University of Toronto

12:00 Lunch – lobby

Graduate Seminar – BSP 130

1:00 "Test of exact solutions for non-EIT fast-light pulses,"
Dave Clader, University of Rochester

1:15 "Femtosecond time-resolved absorption spectroscopy of open chain carotenoids,"
Hong Cong, University of Connecticut

1:30 "Optical coupling and parametric sideband generation in a semiconductor bound exciton,"
Renuka Rajapakse, University of Connecticut

1:45 "Production of highly charged states of molecular nitrogen in an impulsively aligned sample,"
Sarah Nichols, Stony Brook

2:00 "Observation of Lyman-α radiation from femtosecond excitation of hydrogen molecular ions,"
Li Fang and Brad Moser, University of Connecticut

2:15 "Wave-front and wave-packet dissociation in nitrogen molecular ions,"
Ryan Coffee, University of Connecticut

2:30 Break – lobby

Multielectron effects in atoms and molecules – BSP 130

2:45 Introduction
George Gibson, University of Connecticut

3:00 "Electron correlation effects in 1-, 2-, and 3-electron phenomena,"
Chunlei Guo, University of Rochester

3:30 "In-plane theory of non-sequential triple ionization (NSTI),"
Joe Eberly, University of Rochester

4:00 Break – lobby

Ultrafast Applications – BSP 130

4:15 "Ultrafast structural motions during chemical reactions and cancer imaging with partially coherent x-ray sources,"
Christoph Rose-Petruck, Brown University

5:00 "Laser Induced Coincidence Coulomb Imaging,"
Joe Sanderson, University of Waterloo

6:30 Banquet – Nathan Hale Inn

Saturday June 3

Ultracold processes – BSP 130

8:30 TBA,
Juha Javanainen, University of Connecticut

9:00 "Control of ultracold collisions with frequency-chirped light,"
Phil Gould, University of Connecticut

9:30 "BCS-BEC crossover in lattice: A direct diagonalization approach,"
Tun Wang, University of Connecticut

10:00 Break – lobby

Quantum information – BSP 130

10:15 TBA,
Misha Lukin, Harvard University

10:45 "Quantum computing with switchable dipoles,"
Robin Cote, University of Connecticut

11:15 "Negative refraction without absorption using quantum interference,"
Susanne Yelin, University of Connecticut

11:45 Conference ends

Cross Border Workshop On Laser Science, 2006 **University of Connecticut Storrs, CT**

Participant List

Name	Affiliation	Email address
Jean-Paul Brichta	Waterloo	jpobrich@sciborg.uwaterloo.ca
Dave Clader	Rochester	dclader@pas.rochester.edu
Ryan Coffee	UConn	coffee@phys.uconn.edu
Hong Cong	UConn	cong_hong@hotmail.com
Robin Cote	UConn	rcote@phys.uconn.edu
Nirit Dudovich	NRC	Nirit.Dudovich@nrc-cnrc.gc.ca
Joe Eberly	Rochester	eberly@pas.rochester.edu
Li Fang	UConn	lifang@phys.uconn.edu
Harry Frank	UConn	harry.frank@uconn.edu
Oliver Gessner	NRC	Oliver.Gessner@nrc-cnrc.gc.ca
George Gibson	UConn	gibson@phys.uconn.edu
Phil Gould	UConn	plg@phys.uconn.edu
Chunlei Guo	Rochester	guo@optics.rochester.edu
Yifei Huang	UConn	hauangy@phys.uconn.edu
Juha Javanainen	UConn	jj@phys.uconn.edu
Misha Lukin	Harvard	lukin@fas.harvard.edu
Svetlana Malinovskaya	Michigan	smalinov@umich.edu
Brad Moser	UConn	bradley.moser@phys.uconn.edu
Sarah Nichols	Stony Brook	snichols@grad.physics.sunysb.edu
Matt Partlow	Toronto	mpartlow@physics.utoronto.ca
Brett Pearson	Stony Brook	bpearson@ms.cc.sunysb.edu
Renuka Rajapakse	UConn	rajapaks@phys.uconn.edu
Christoph Rose-Petruck	Brown	Christoph_Rose-Petruck@brown.edu
Joseph Sanderson	Waterloo	j3sander@scimail.uwaterloo.ca
Helmut Satzger	NRC	satzger@ned.sims.nrc.ca
Win Smith	Uconn	winthrop.smith@uconn.edu
William Stwalley	UConn	w.stwalley@uconn.edu
Ben Sussman	NRC	ben.sussman@nrc.ca
Fraser Turner	Waterloo	fraserct@gmail.com
Tun Wang	UConn	tun.wang@uconn.edu
Tom Weinacht	Stony Brook	tweinacht@ms.cc.sunysb.edu
Neal Woodbury	Arizona	NWOODBURY@asu.edu
Susanne Yelin	UConn	susanne.yelin@uconn.edu
Lu Zhao	UConn	zhaol@phys.uconn.edu

Test of Exact Solutions for Non-EIT Fast-Light Pulses

B.D. Clader*, Q-Han Park and J.H. Eberly
Department of Physics and Astronomy,
University of Rochester, Rochester, NY 14627 USA

We provide exact analytic solutions for short pulses in finite-length media and use them to suggest numerical tests whether the phenomna of so-called fast light and slow light can be extended into the short-pulse domain. In the example presented here we restrict attention to short-pulse fast light. We consider two-level media subject to dephasing arising from inhomogeneous broadening. We write a state vector for a typical medium atom in the form $|\psi(x,t)\rangle = c_1(x,t)|1\rangle + c_2(x,t)|2\rangle$, and look at the coupled Maxwell-Schrödinger equations in the slowly varying envelope approximation which are

$$\frac{\partial c_1}{\partial t} = i\frac{\Omega}{2}c_2, \qquad \frac{\partial c_2}{\partial t} = i\frac{\Omega}{2}c_1 - i\Delta c_2, \quad \text{and} \quad \left(c\frac{\partial}{\partial x} + \frac{\partial}{\partial t}\right)\Omega = -ig\langle c_1 c_2^*\rangle, \tag{1}$$

where $\Omega = 2\mu E/\hbar$ is the Rabi frequency associated with the $1 \to 2$ transition and for simplicity is assumed to be real, $\Delta = \omega_{21} - \omega$ is the detuning of the laser from the transition frequency ω_{21}, μ is the dipole moment matrix element, and E is the electric field envelope function. The $\langle \dots \rangle$ indicates an average over an inhomogeneous distribution of detunings with lifetime T_2^*, $g = N\mu^2\omega/\epsilon_0\hbar$ is the atom-field coupling parameter, N is the density of resonant atoms, and ω is the probe pulse frequency. Note that the absence of relaxation terms for the atoms must be supported self-consistently by the Maxwell equation in the sense that the only useful solutions for Ω will be those pulses short enough for medium relaxation to be assumed ineffective.

Superluminal propagation occurs in amplifiers[1, 2], so for the simplest demonstration we prepare a medium in its upper level and inject a pulse substantially shorter than the upper-level lifetime. We are concerned with the practical case of a finite medium located between the positions x_0 and x_1, and with pulse shapes that vary during propagation. Very simple exact fast-pulse solutions are given by the following segmented expression:

$$\Omega(x,t) = \begin{cases} \frac{2}{\tau}\mathrm{sech}\frac{1}{\tau}\left(t - \frac{x}{c}\right) & x < x_0 \\ \frac{2}{\tau}\mathrm{sech}\frac{1}{\tau}\left(t - \frac{x}{v_g} + \phi_0\right) & x_0 \leq x \leq x_1 \\ \frac{2}{\tau}\mathrm{sech}\frac{1}{\tau}\left(t - \frac{x}{c} + \phi_1\right) & x > x_1, \end{cases} \tag{2}$$

where $\phi_0 = -\tau^2 g x_0/2c$, and $\phi_1 = \tau^2 g(x_1 - x_0)/2c$, and τ is obviously the probe pulse duration. The group velocity of the pulse in the absence of inhomogeneous broadening is given by $v_g/c = 1/(1 - g\tau^2/2)$. One can obtain either $v_g > c$ or $v_g < 0$ depending on $g\tau^2$.

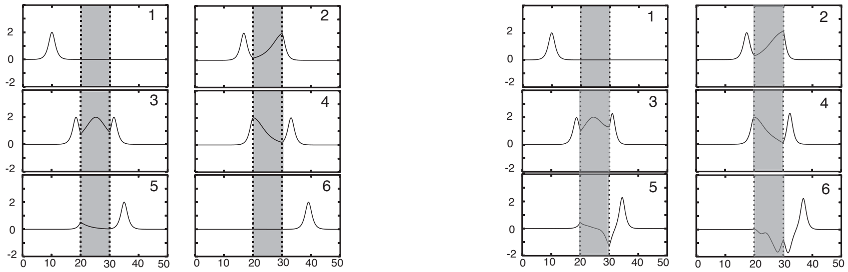

FIG. 1: Snapshots of analytic and numerical fast-light pulse propagation. The horizontal x axes are in units of $c\tau$. The shaded region indicates the location of the medium. We have chosen parameters such that $v_g/c \approx -3.27$. Left figure: Analytic solutions, as given in Eq. (2). Note in frame 2 the emergence at $x = x_1 = 30$ of the greatly advanced peak of the pulse. Right figure: Numerical solution to Eqns. (1) with the same initial pulse shape but for a pulse initially entirely outside the medium with abrupt edges at ±25 units from the peak.

We will discuss the relation of the unphysical exact sech solutions to numerical solutions using pulses with finite tails. Snapshots of both are shown in Fig. 1.

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[1] N. G. Basov *et. al.*, *"Nonlinear Amplification of Light Pulses,"* Sov. Phys. JETP 23, 16-22 (1966).
[2] A. Icsevgi and W. E. Lamb, Jr., *"Propagation of Light Pulses in a Laser Amplifier,"* Phys. Rev. 185, 517-545 (1969).

FEMTOSECOND TIME-RESOLVED ABSORPTION SPECTROSCOPY OF OPEN CHAIN CAROTENOIDS

Hong Cong[2], Dariusz Niedzwiedzki[1], George N. Gibson[2] and Harry A. Frank[1]
[1] Department of Chemistry, University of Connecticut, Storrs, CT 06269-3060, USA;
[2] Department of Physics, University of Connecticut, Storrs, CT 06269-3046, USA.

Abstract

Many of the spectroscopic features and photophysical properties of carotenoids are explained using a three-state model where the strong visible absorption of the molecules is associated with an S_0 ($1^1A_g^-$) → S_2 ($1^1B_u^+$) transition, and the lowest lying singlet state, S_1 ($2^1A_g^-$), is a state into which absorption from the ground state is forbidden by symmetry. However, semi-empirical and *ab-initio* quantum calculations have suggested that additional excited singlet states may lie between S_1 and S_2, and spectroscopists have reported evidence for these states using ultrafast laser methodologies. One such state, denoted S*, was invoked by van Grondelle and coworkers[1] to account for the ultrafast dynamics of the carotenoid, spirilloxanthin, in solution and in the LH1 complex from *Rhodospirillum rubrum* being different at different probe wavelengths, and in the LH1 complex leading to triplet state formation. Initially, S* was thought to be formed only in the very long (N=13) spirilloxanthin molecule. However subsequent studies on β-carotene, lycopene, rhodopin glucoside, zeaxanthin and spheroidene have suggested that S* may occur more commonly. The primary spectroscopic characteristics of S* are that it has a broad transition with a maximum in the wavelength region between the $S_0 \rightarrow S_2$ and $S_1 \rightarrow S_n$ absorption bands, and that it decays in several picoseconds. An alternative view of the origin of S* has been published by Wohlleben et al[2] who argue that, in solution, S* is a vibrationally-excited, "hot" ground state populated by a combination of impulsive Raman scattering of the $S_0 \rightarrow S_2$ pump pulse and internal conversion from S_1. In this work we present the results of a systematic, ultrafast, time-resolved spectroscopic investigation of *cis* and *trans* geometric isomers of HPLC-purified spirilloxanthin (N=13), rhodopin glucoside (N=11), and spheroidene (N=10) in acetone and CS_2 solutions at room temperature and in EPA (5:5:2 v/v/v ether: isopentane: ethanol) at 77 K. Analysis of the data using global fitting techniques reveals the inherent spectral properties and ultrafast dynamics of each of these molecules and addresses the structural features controlling S* formation in solution.

Objectives

- To examine the spectroscopic properties of open-chain carotenoids
- To measure the ultrafast dynamics of their excited electronic states
- To elucidate the factors controlling the formation and decay of S*

Conclusions

- S* is an excited state of the carotenoid having a twisted conformation.
- S* is formed in high yield in solvents such as CS2 that promote conformational disorder.
- The yield of S* does not depend on whether the molecule is initially in an all-trans or central-cis geometric configuration.

References

(1) Gradinaru, C. C.; Kennis, J. T. M.; Papagiannakis, E.; van Stokkum, I. H. M.; Cogdell, R. J.; Fleming, G. R.; Niederman, R. A.; van Grondelle, R. *Proc. Natl. Acad. Sci. U.S.A* **2001**, *98*, 2364.

(2) Wohlleben, W.; Buckup, T.; Hashimoto, H.; Cogdell, R. J.; Herek, J. L.; Motzkus, M. ; *J. Phys. Chem. B* **2004**, *108*, 3320.

Femtosecond Coherent Control for Nonlinear Spectroscopy and microscopy

Nirit Dudovich

NRC, Ottawa, Canada

Femtosecond pulses are the main tool of nonlinear spectroscopy. However, inducing molecular or atomic transitions by femtosecond pulses typically results in reduced spectral resolution due to the broad spectral bandwidth of the pulse. I will present various quantum control methods by which the spectral resolution is restored. Investigation of the basic interaction between broadband pulses and narrow band transitions in the perturbative regime has led to the development of various schemes to manipulate the outcome of such interactions. These schemes were applied to manipulate coherent Raman spectroscopy induced by a single pulse. In the presentation I will describe how by controlling the pulse shape we achieved high spectral resolution, nearly two orders of magnitude higher than the pulse bandwidth. High spectral resolution CARS process using a single broadband laser source is particularly attractive for nonlinear microscopy applications as we have demonstrated by constructing a CARS microscope operating with a single laser beam. The concept of performing a nonlinear optical interaction in a single coherently controlled pulse offers a promising alternative to standard multiple-pulse nonlinear systems in use today.

In-plane Theory of Non-Sequential Triple Ionization (NSTI)

Phay J. Ho and J. H. Eberly
Department of Physics and Astronomy, University of Rochester, NY 14627, USA.

Abstract: We describe, and show in video format, the results of first-principles in-plane numerical calculations of non-sequential triple ionization (NSTI) of atoms in a linearly polarized intense laser pulse. In a fully classically correlated description, all three electrons respond dynamically to the nuclear attraction, the pairwise e-e repulsions and the laser force throughout the duration of a 780nm laser pulse. Non-sequential ejection is shown to occur in a multi-electron, possibly multi-cycle and multi-dimensional, rescattering sequence that is coordinated by a number of sharp transverse recollimation impacts.

Several recent experiments have begun to probe the process of correlated-electron intense-field multiple ionization of atoms beyond two electrons [1]. Theoretical investigation of this process has barely begun [2] and any study using a fully quantum treatment will be substantially hindered by the necessity to include multiple-electron interactions. However, motivated by the success of a two-electron classical theory [3] in capturing the experimental features of double ionization, we extend the theory to include a third electron and explore the mechanism of intense-field triple ionization.

In this 3e classical study, 1 million classical 3e trajectories are used to obtain the non-perturbative response of a 3e system exposed to a linearly polarized laser pulse. In our study, we abandon the aligned electron approximation and extend the classical method to a plane that contains the polarization (x) axis and one transverse (y) axis. This extension allows us to illuminate the role of the nucleus clearly along the transverse axis.

The calculations are done using different 20fs 780nm laser pulses with laser intensities in the range of 10^{14} W/cm^2 to 10^{16} W/cm^2. At the end of the pulse, we determine the fraction of the 3e trajectories that are triply ionized as a function of intensity. A theoretical "knee" structure is obtained for the triple ionization ion count. The "knee" is an indicator of strong e-e correlation in the process of triple ionization. We verify the existence of the multi-electron correlation by examining the history of the chaotic triple ionized trajectories. Despite the chaotic nature of the periodically driven non-linear interaction, a complex recollision mechanism is coherently displayed among the pathways to triple ionization.

In the cases where the recolliding electron undergoes transverse displacement, a universal process of transverse collimation is observed. Rather than a continuous pull, the nuclear force affects the electron with a series of weak impulses and these impulses produce step-like jumps in the transverse momentum. The timings of these impulses coincide with the times that the recolliding electron crosses the nucleus (i.e. x=0).

Acknowledgement: Support has been provided through DOE grant DE-FG02-05ER15713.

References:

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Direct femtosecond laser excitation of the 2p state of H by a resonant 7-photon transition in H_2^+

Li Fang and Brad Moser
University of Connecticut, Storrs, CT

Experiments have shown that high rates of multiphoton transition of molecules to dissociative energy levels leaving atoms in highly excited electronic states can be achieved [1][2]. Recently, a new theory of three-level systems predicts much higher multiphoton transition rates than that predicted by theory of traditional two-level systems [3][4]. In the current work, two independent experiments on hydrogen, vacuum ultraviolet (VUV) and time of flight (TOF), shows a more than six-photon excitation of two near degenerate excited electronic states (i.e., $2s\sigma_g$ and $3p\sigma_u$) for which the dissociative fragment H is in the 2p state. In the VUV experiments, the fluorescent signal from a single 25fs laser pulse demonstrates the production of Lyman α radiation which represents transition from 2p atomic state to 1s atomic state. Using the pump-probe technique with VUV, depletion of radiation as probe-laser intensity increases is observed, which is due to ionization of neutral atoms by the probe pulse. In the TOF pump-probe experiments, dissociative fragment H+ as a product of this ionization is detected. Corresponding to depletion of Lyman α radiation in VUV, enhancement of H+ signal as a function of probe intensity is observed. Moreover, making use of the different ionization thresholds of the 2s and 2p atomic states, production rate for each state can be quantified.

[1] John P. Nibarger, Ming Li, Saipriya Menon, and George N. Gibson, PRL 83, 4975 (1998).
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Femtosecond Multidimensional Imaging - Watching Chemistry from the Molecule's Point of View

Oliver Gessner

Steacie Institute for Molecular Sciences, Ottawa, Canada

Observing single molecules undergoing chemical reactions in real-time has provided fundamental progress in the understanding of reaction dynamics on a microscopic level. The two basic components for making a "molecular movie" are the same as those needed to capture dynamic processes in the macroscopic world: A light source or shutter providing an exposure time which is short compared to the timescale of the dynamics and an imaging system with sufficient resolution to uncover the changes between consecutive exposures. The combination of femtosecond lasers and photoelectron spectroscopy is a well proven method to observe ultrafast molecular dynamics in the gas phase. The technique, termed time-resolved photoelectron spectroscopy (TRPES), is sensitive to both electronic and vibrational dynamics and is in principle not affected by the presence of any dark states. However, chemistry takes place in the reference frame of the molecule while TRPES measurements are made in the lab frame averaging over all orientations in space of the free molecules. The loss of information about the molecular orientation potentially leads to a loss of physical information in the experiment. By applying a new technique, time-resolved coincidence imaging spectroscopy (TRCIS), we eliminate this problem and for the first time are able to record a molecular movie from the molecule's point of view [1]. The combination of TRPES and TRCIS enables us to follow the entire dissociation pathway of the UV-excited NO dimer all the way from the initial excitation to the final product emission. A complex dissociation mechanism is revealed involving intermediate configurations which, corroborated by high level ab initio calculations, are characterized during all stages of the reaction. Furthermore, we directly observe the evolution of intramolecular vibrational energy redistribution (IVR) during the dissociation by measuring time-dependent product state distributions. The study constitutes a showcase example for the new level of insight into charge- and energy-flow during chemical reactions provided by time-resolved multidimensional imaging methods.

[1] O. Gessner *et al.*, Science 311, 219 (2006); published online 15 December 2005 (10.1126/science.1120779).

Ultrafast Coherent Spectroscopy of Biological Systems

Svetlana Malinovskaya

Department of Physics, University of Michigan
Ann Arbor, MI 48109

Advancements in coherent control of vibrational dynamics lie in the development of novel noninvasive imaging techniques that provide information about biological species with molecular specificity. Of particular interest is development of coherent Stokes and anti-Stokes Raman scattering (CSRS and CARS) spectroscopies, which contain vibrational finger-prints of molecules. CSRS and CARS are nonlinear optical processes utilizing ultrafast laser pulses to generate strong, molecular-specific signals that can be used for molecular detection. Imaging techniques based on the CSRS and CARS use selective excitation of Raman transitions at predetermined frequencies and involve ultrafast laser pulse manipulation. Ultrafast pulse shaping and the use of chirped delayed pulses will be discussed as the means of vibrational mode selective excitation in impulsive and non-impulsive Raman scattering. The role of the intramolecular processes, such as the coupling between Raman vibrational modes, that occur on a femtosecond time scale and affect the ultrafast energy redistribution will be revealed. Liquid phase aspects of coherent control techniques arising from the molecular interaction with the solvent environment will be also discussed.

Production of Highly Charged States of Molecular Nitrogen in an Impulsively Aligned Sample

Sarah R. Nichols, **Brett J. Pearson**, **and Thomas C. Weinacht**
Department of Physics and Astronomy, Stony Brook University, Stony Brook, New York 11794-3800 USA

Abstract: We examine molecular dynamics in ion fragments resulting from multiply charged states of molecular nitrogen. Preliminary measurements of our time-of-flight mass spectra show that the production of energetic fragments from multiply ionized molecules is significantly affected by the dynamics of an impulsively created rotational wavepacket.

Summary: Nitrogen molecules are a useful simple system for investigating molecular dynamics. As a simple rigid rotor system, the strong field ionization of aligned nitrogen can be both modeled and measured relatively simply.[1] These aligned molecules are useful in high-order harmonic generation[2] and can be imaged using traditional time-of-flight as well as ion imaging techniques.[1,3] High resolution time-of-flight systems, as well as the use of correlation matrices, permit the assignment of ion fragment subpeaks to different dissociation channels, where the kinetic energy of dissociation causes fragments to arrive “early” or “late” for their mass.[4] We report the observation of molecular alignment not only in the main ion fragments of N_2^+, as expected, but also through energetic N^+ fragments, which come from higher ionization states. In addition, we see alignment in the N^{2+} peaks (7 amu), which can only come from multiply ionized states (N_2^{2+} and higher). We are also investigating alignment in larger molecules such as 1,1,1-trifluoroacetone, CH_3COCF_3.

References:

[1] I.V. Litvinyuk, K.F. Lee, P.W. Dooley, D.M. Rayner, D.M. Villeneuve, and P.B. Corkum, “Alignment-Dependent Strong Field Ionization of Molecules,” Phys. Rev. Lett. **90** 233003 (2003).
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Quantum Tomography of Cold Atomic Systems *

Matthew Partlow
Centre for Quantum Information & Quantum Control and
Institute for Optical Sciences
Department of Physics, University of Toronto, Canada

June 2nd, 2006

The development of the fields of quantum information and quantum computation has necessitated a more pragmatic investigation into the measurement and control of quantum systems. Experiments with trapped cold atoms provide an excellent means of understanding key issues in quantum coherence, including the characterization and control of decoherence within real-world constraints.

In our optical lattice system we have explored several techniques of measurement and control using a limited set of lattice manipulations. With a shallow lattice supporting only two bound states, we are able to selectively prepare quantum vibrational states and directly measure their phase-space distribution as well as their 2×2 density matrix. Obtaining a complete set of density matrices, measured before and after a given process, yields complete characterization of the state evolution due to that process. This "Quantum Process Tomography" (QPT) is used to characterize the resonant coupling and decoherence within the system. Furthermore, we have used this technique to optimize a pulse echo sequence in order to preserve coherence. Currently, to distinguish between homogeneous and inhomogenous effects of decoherence, we are developing a technique for 2D spectroscopy of coherent state oscillations.

Additionally, on our BEC apparatus, we are developing a possible tool for performing phase-space tomography of an expanding condensate. The technique relies on the scattering of the atomic cloud off of a repulsive barrier, which can be modified during the collision, and measuring the interference between scattered and non-scattered momentum components.

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[*] Samansa Maneshi, Jalani Kanem, Chao Zhuang, Mirco Siercke, Chris Ellenor, Rockson Chang, and Aephraim Steinberg

Dissociative Wave Packets in Large Molecules: Control and Measurement

Brett J. Pearson[1], David Cardoza[1], Carlos Trallero[1], Mark Baertschy[2], and Thomas C. Weinacht[1]
[1] *Department of Physics and Astronomy, Stony Brook University, Stony Brook, New York 11794-3800 USA*
[2] *Department of Physics, University of Colorado at Denver, Denver, Colorado 80217 USA*

Abstract: Ultrafast control experiments in a polyatomic molecule uncover a dynamic resonance between different electronic states during dissociation of the molecular ion. Using this resonance allows one to map out the amplitude and phase of the nuclear wave function along the dissociation coordinate. The technique is general and can be used in molecules that undergo dynamic resonances during dissociation and whose products are distinguishable via time-of-flight mass spectrometry.

Summary: We interpret a series of learning control experiments on ionization and fragmentation in halogenated acetone molecules using electronic structure calculations and pump-probe spectroscopy. A common feature in the experiments is a dynamic charge redistribution during molecular dissociation.

The results are understood using a wave packet description on ionic potential energy surfaces (PES); an initial laser pulse promotes population onto a dissociative PES, while a subsequent probe pulse couples this wave packet to a second dissociative PES via the dynamic resonance. The charge transfer process, initiated by a time-delayed probe pulse, serves to measure the probability density of the dissociating wave packet as a function of time at the resonance location [1].

While a single-pulse, pump-probe experiment measures $|\Psi(r_0,t)|^2$, using a double-pulse creates two wave packets that interfere on the excited PES. This inference provides phase information about the wave function along the dissociation coordinate [2-4].

References:

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Optical coupling and parametric sideband generation in a semiconductor bound exciton

Renuka Rajapakse

University of Connecticut, Storrs, CT

Group theoretical techniques are used to deduce the selection rules and energy splitting of the electric dipole absorption lines $\Gamma_6 \rightarrow \Gamma_8$, $\Gamma_7 \rightarrow \Gamma_8$ of a donor exciton in a tetrahedral semiconductor, e.g., GaAs. We obtain selection rules for the above transitions for the spin states m_j. The application in a bound exciton system in a magnetic field for the purposes of obtaining electromagnetically induced transparency is discussed. In particular, Stokes and Anti Stokes couplings have been experimentally observed in such a system. We theoretically calculate the expected gains of the Stokes and anti Stokes couplings for σ- and π-polarization of the pump field. We show that the system can be interpreted as one exhibiting double Λ-type transitions, and therefore could be used for coherent non-linear optics and, ultimately, quantum-optics based quantum information processing.

Ultrafast structural motions during chemical reactions and cancer imaging with partially coherent x-ray sources

Christoph Rose-Petruck

Brown University, Providence, Rhode Island

The talk presents our latest results in various research areas:

1. Picosecond time-resolved structure measurements during chemical reactions using an ultrafast laser-driven x-ray source
2. Structural deformations of iron pentacarbonyl in aromatic and protic solvents and their implications for ultrafast ligand substitution reactions
3. High-resolution and high-contrast medical imaging of cancer in soft tissue using partially coherent x-ray sources in combination with the simultaneous application of ultrasound waves

Details:

1. Ultrafast high-intensity laser pulses incident upon condensed matter targets can generate high-density plasmas that emit x-ray pulses with sub-picosecond temporal structure, significant spatial coherence, and high brightness. Such laser-driven plasma x-ray sources operating at kilohertz repetition rates have been developed in our laboratory and are used for structural measurements of various solvated transition metal complexes by XAFS spectroscopy. Specifically, picosecond laser-pump XAFS-probe measurements of $Fe(CO)_6^{4-}$ in water have been performed at various pump-probe delay times before and after UV-photoexcitation of the complex. Picosecond time-resolved XAFS data are presented and discussed.

2. Most ultrafast chemical processes studied to date are primarily unimolecular. Bi-molecular processes may subsequently occur thought the diffusive encounter of excited molecules and other reactants. These diffusive processes proceed within picoseconds at which time possible photo induced coherent motions have been randomized. Since it would be desirable to observe bi-molecular reaction without the influence of uncontrollable diffusions, we attempt to address this problem by “pre-assembling” reactant complexes in solutions before photoexcitation. In many organic solvents, $Fe(CO)_5$ exhibits substantial deformations from its gas-phase D_{3h} symmetry. These deformations are accompanied by the association of $Fe(CO)_5$ with <u>one</u> solvent molecule forming a complex that, in principle, permits ultrafast ligand substitution reactions possibly before the onset of decoherence.

3. Conventional x-ray imaging relies on the differential absorption of x-radiation in tissues. This leads to low image contrast in soft tissue. Our imaging method relies on the differential phase-shift of x-rays causing image contrast even in the absence of any x-ray absorption. This concept is similar to optical in-line holography but is done with ~ 50-keV x-rays. Example images of tumor-infiltrated livers in a murine model (Female BALB/c (H-2d) are presented and discussed. The tumors are induced by injection of tumor cells in the spleen or liver. A further refinement of this imaging method relies on ultrasound waves simultaneously applied to the tissues during x-ray imaging. Due to the tissues’ differential acoustic impedance, differential movements are induced that can be detected with high contrast.

Laser Induced Coincidence Coulomb Imaging

J. H. Sanderson, J.P. Brichta, S. Walker, X. Sun

Department of Physics and Astronomy University of Waterloo

Laser induced Coulomb imaging is a promising technique for examining molecular dynamics and understanding the influence on molecules in an intense laser field. In the long pulse (500-100fs) regime the technique is able to diagnose the effect of laser induced excitation and dissociation while at the same time resolving intrinsic dynamics such as the competition between concerted and sequential unimolecular reactions [1]. With the advent of few cycle pulses (<10fs) Coulomb images are approaching equilibrium molecular structure in terms of bond angles while, bond length imaging is still problematic [2]. Few cycle pulses have also allowed the first images of nuclear dynamics to be made [3]. We will describe the setup at Waterloo for measuring molecular geometry and dynamics, which consists of a new stretcherless regenerative amplifier laser system, a hollow fiber compression setup and a time and position sensitive ion coincidence apparatus. We will review progress to date in achieving high temporal and spatial resolution images, discuss the issues of current interest such as the influence of ionization dynamics on the imaging process and consider future directions.

Ref

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2. Légaré F *et al* Phys. Rev.A **71**, 013415 2005
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Parametric Multi-Frequency Raman Generation

F. C. Turner, A. Trottier, D. Strickland
Department of Physics, Guelph-Waterloo Physics Institute, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada
fcturner@uwaterloo.ca

L. L. Losev
Division of Quantum Radio Physics, P.N. Lebedev Physical Institute, Leninsky Prospekt 53, 119991 Moscow, Russia

Abstract: High-order stimulated Raman scattering was used to generate coherent radiation from 1.49μm to 312nm. Its scaling with intensity demonstrated that energy was preferentially pumped into the Anti-Stokes side of the spectrum, and by the final intensity at least three Anti-Stokes orders were as intense as the pump beam itself.

The effort to produce ultra-short pulses relies on the coherent generation of a broad range of optical frequencies. To obtain this large bandwidth, one can take advantage of parametric stimulated Raman scattering to produce high orders of Stokes and anti-Stokes radiation [1]. This technique of multi-frequency Raman generation (MRG), involves irradiating a medium by a pump beam containing frequency components that match a particular Raman transition. The radiation couples the ground and excited state and simultaneously scatters into its corresponding Stokes and anti-Stokes frequencies. This process cascades as the pump, Stokes and anti-Stokes frequencies again couple the two states and scatter into higher orders.

By focussing our Raman pump beams into a 250μm hollow glass fibre filled with SF_6 we were able to detect 32 Raman sidebands with a wavelength range from 1.49μm down to 312nm. Using a variable attenuator, we investigated the scaling of this process with increasing pump energy. We determined that energy is pumped preferentially in to the shorter-wavelength Anti-Stokes orders, in such a way as to form a plateau with intensities comparable to the transmitted pump intensity. This growth continued until the onset of the competing non-linear process of Self-Phase Modulation. With the proper phase compensation, the generated spectra have the potential to produce a central pulse with a FWHM of 3fs with an estimated 140μJ of energy.

[1] L. L. Losev and A. P. Lutsenko, “Ultrabroadband parametric stimulated Raman scattering in a highly transient regime,” Opt. Comm. **132**, 489-493

BCS-BEC crossover in lattice: A direct diagonalization study

T. Wang, J. Javanainen, and S. F. Yelin

University of Connecticut, Storrs, CT

BCS - BEC crossover through Feshbach resonance of atomic Fermi gases in one dimensional optical lattice is studied with a direct diagonalization approach. It is shown that in the crossover regime, the molecular bosons as well as the fermions have strong fluctuation and they are strongly correlated. The order parameters and thus the critical temperature for the superfluid transition are enhanced in the crossover regime. Further more, BCS superfluidity can exist close to the full filling due to the boson-fermion coupling term.