



Defense Threat Reduction Agency  
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DTRA-TR-12-65

# TECHNICAL REPORT

## Manipulation of Molecular Quantum Wavepackets with Ultrashort Laser Pulses for Non-destructive Detection of Volatile Explosives

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February 2013

HDTRA1-09-1-0021

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14. ABSTRACT Experimental demonstration of the ability to control Raman excitation of simple molecular systems in the strong field regime. Assessing the feasibility of applying the strong-field control techniques to more complex molecular systems. Experimental and theoretical development of alternative approaches to molecular identification with ultrashort laser pulses.					
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# CONVERSION TABLE

Conversion Factors for U.S. Customary to metric (SI) units of measurement.

MULTIPLY → BY → TO GET  
TO GET ← BY ← DIVIDE

angstrom	1.000 000 x E -10	meters (m)
atmosphere (normal)	1.013 25 x E +2	kilo pascal (kPa)
bar	1.000 000 x E +2	kilo pascal (kPa)
barn	1.000 000 x E -28	meter <sup>2</sup> (m <sup>2</sup> )
British thermal unit (thermochemical)	1.054 350 x E +3	joule (J)
calorie (thermochemical)	4.184 000	joule (J)
cal (thermochemical/cm <sup>2</sup> )	4.184 000 x E -2	mega joule/m <sup>2</sup> (MJ/m <sup>2</sup> )
curie	3.700 000 x E +1	*giga bacquerel (GBq)
degree (angle)	1.745 329 x E -2	radian (rad)
degree Fahrenheit	$t_k = (t^{\circ}f + 459.67) / 1.8$	degree kelvin (K)
electron volt	1.602 19 x E -19	joule (J)
erg	1.000 000 x E -7	joule (J)
erg/second	1.000 000 x E -7	watt (W)
foot	3.048 000 x E -1	meter (m)
foot-pound-force	1.355 818	joule (J)
gallon (U.S. liquid)	3.785 412 x E -3	meter <sup>3</sup> (m <sup>3</sup> )
inch	2.540 000 x E -2	meter (m)
jerk	1.000 000 x E +9	joule (J)
joule/kilogram (J/kg) radiation dose absorbed	1.000 000	Gray (Gy)
kilotons	4.183	terajoules
kip (1000 lbf)	4.448 222 x E +3	newton (N)
kip/inch <sup>2</sup> (ksi)	6.894 757 x E +3	kilo pascal (kPa)
ktap	1.000 000 x E +2	newton-second/m <sup>2</sup> (N-s/m <sup>2</sup> )
micron	1.000 000 x E -6	meter (m)
mil	2.540 000 x E -5	meter (m)
mile (international)	1.609 344 x E +3	meter (m)
ounce	2.834 952 x E -2	kilogram (kg)
pound-force (lbs avoirdupois)	4.448 222	newton (N)
pound-force inch	1.129 848 x E -1	newton-meter (N-m)
pound-force/inch	1.751 268 x E +2	newton/meter (N/m)
pound-force/foot <sup>2</sup>	4.788 026 x E -2	kilo pascal (kPa)
pound-force/inch <sup>2</sup> (psi)	6.894 757	kilo pascal (kPa)
pound-mass (lbm avoirdupois)	4.535 924 x E -1	kilogram (kg)
pound-mass-foot <sup>2</sup> (moment of inertia)	4.214 011 x E -2	kilogram-meter <sup>2</sup> (kg-m <sup>2</sup> )
pound-mass/foot <sup>3</sup>	1.601 846 x E +1	kilogram-meter <sup>3</sup> (kg/m <sup>3</sup> )
rad (radiation dose absorbed)	1.000 000 x E -2	**Gray (Gy)
roentgen	2.579 760 x E -4	coulomb/kilogram (C/kg)
shake	1.000 000 x E -8	second (s)
slug	1.459 390 x E +1	kilogram (kg)
torr (mm Hg, 0° C)	1.333 22 x E -1	kilo pascal (kPa)

\*The bacquerel (Bq) is the SI unit of radioactivity; 1 Bq = 1 event/s.

\*\*The Gray (GY) is the SI unit of absorbed radiation.

## Final Report from the University of British Columbia (Investigators: Milner, Shapiro)

### ii) Objectives:

Experimental demonstration of the ability to control Raman excitation of simple molecular systems in the strong field regime. Assessing the feasibility of applying the strong-field control techniques to more complex molecular systems. Experimental and theoretical development of alternative approaches to molecular identification with ultrashort laser pulses.

### iii) Status of effort:

The work on studying the prospects of strong-field coherent control in simple molecules (molecular Iodine) has been completed, both on the side of experimental demonstration and theoretical analysis. In addition, five new alternative approaches to remote molecular detection with ultrafast coherent optical processes have been successfully developed and investigated: (1) interferometric coherent anti-Stokes Raman scattering with noisy laser pulses; (2) coherent nonlinear spectroscopy in scattering materials; (3) time-resolved ro-vibrational spectroscopy; (4) polarization modulation spectroscopy with coherent anti-Stokes scattering; and (5) spectroscopic detection with electromagnetically induced transparency.

### iv) Accomplishments/New Findings:

**Strong-field control of CARS:** Strong-field effects have been detected as saturation of coherent anti-Stokes Raman scattering signal with increasing pulse energies (exceeding  $10^{13}$  W/cm<sup>2</sup>). We have implemented a number of schemes to avoid the detrimental effect of ro-vibrational “ladder climbing” – i.e. the re-distribution of molecules among multiple ro-vibrational levels, which effectively bleaches the sample and results in the saturation of the CARS signal. The effects of two pulse shaping techniques, namely (a) frequency chirping and (b) breaking a single excitation pulse into a train of pulses, have been investigated both theoretically and experimentally. From the comparison of these two control mechanisms, we have found that applying a frequency chirp is more effective in suppressing the undesired ladder climbing (and therefore heating the molecules), whereas pulse trains are useful in enhancing the overall CARS signal in the strong-field limit.

**Interferometric CARS with noisy fields:** We proposed and implemented a new technique for comparing two Raman active samples. The method employs optical interference of the signals generated via coherent anti-Stokes Raman scattering (CARS) of broadband laser pulses with noisy spectra. It does not require spectrally resolved detection, and no prior knowledge about either the Raman spectrum of the samples or the spectrum of the incident light is needed. We

have studied the proposed method theoretically and also demonstrated it in a proof-of-principle experiment on toluene and ortho-xylene samples.

**Coherent nonlinear spectroscopy in scattering materials:** We studied the prospects of controlling transmission of broadband laser pulses through turbid samples for the purpose of chemical detection with nonlinear optical methods. The ability to focus transmitted broadband light is limited via both the scattering properties of the medium, and the technical characteristics of the experimental set-up. We have identified two time scales, given by pulse stretching in the near- and far-field regions, which define the maximum bandwidth of a pulse amenable to focusing. Experimental work is underway to verify our theoretical findings and demonstrate the method with real scattering materials.

**Time-resolved ro-vibrational spectroscopy:** We performed an experimental and theoretical study of the evolution of vibrational coherence in a thermal ensemble of nitrogen molecules. Rotational dephasing and rephasing, commonly faced as a major source of vibrational decoherence, was detected by coherent anti-Stokes Raman scattering. The existence of ro-vibrational coupling, which stems from the centrifugal distortion of molecular bonds, and the discrete energy spectrum of the rotational bath lead to a whole new class of full and fractional ro-vibrational revivals – a complex dephasing/rephasing temporal profile of the observed CARS signal. This oscillatory temporal profile is as unique to a particular molecule as its vibrational or rotational spectrum alone, and as such can be used for molecular identification. Following the rich ro-vibrational dynamics on a nanosecond time scale with sub-picosecond time resolution, we were able to determine the second-order ro-vibrational constant and assess new possibilities of controlling decoherence.

**Polarization modulation spectroscopy with coherent anti-Stokes Raman scattering:** We proposed and developed a new technique of coherent anti-Stokes Raman spectroscopy (CARS) with a single femtosecond laser beam. Our approach combines, for the first time, the advantages of the method of spectral pulse shaping with the technique of fast polarization modulation. The former offers good spectral resolution and flexibility to match the spectral profile of the excitation pulses to the particular vibrational dynamics of the molecules of interest, whereas the latter enables one to improve the detection sensitivity, as well as to probe the Raman depolarization factor of the molecular vibration in addition to the traditionally measured vibration frequency.

**Spectroscopic detection with electromagnetically induced transparency:** We studied theoretically the absorption characteristics of atoms and molecules under the condition of Electromagnetically Induced Transparency (EIT). We refer to this research question in two ways which complete the overall understanding of the EIT phenomenon. The first approach is the dynamical approach, which means that we describe the decay process as an interaction with a continuum set of quantum states, and we solve the Time Dependent Schrodinger Equation (TDSE), in order to follow the transient behavior of the decay process. The results of this approach were published in [5], and they reveal few important characteristics of EIT absorption.

We predict how the absorption spectrum will look during the pulse time and at the asymptotic time  $t=\infty$ . Moreover, we predict that pulses will always experience absorption, although the latter is reduced in comparison with the regular absorption (without EIT). We also predict the absorption spectrum during the propagation of the field in molecular medium, and in dynamical pulse sequencing such as stored light process. The knowledge that we obtained from this work can be exploited in fast spectroscopic measurement, using pulses within EIT.

The second approach is stationary, and it deals with the ability to decipher complicated spectra. In this work we used the theoretical formalism of scattering resonances to analyze the necessary conditions for observation of overlapping transition lines in obtained spectrum. We considered two scenarios in which the problem of overlapping transition lines is found. The two scenarios are inter- and intra-molecular overlaps between spectroscopic transition lines. The first scenario points to the problem of a spectrum of small minority of absorbers being hidden by large majority of other absorbers. The second scenario points to the case when the spectrum of a single absorber has overlapping transition lines which make it difficult to decipher the obtained spectrum.

Our proposed method to unravel these problems is to employ the EIT phenomenon in the spectroscopic measurements. The logic behind it is given as follows. First, we recognize the suspected line, i.e. lines that are not predicted in theory, or that suffer from some unpredictable broadening and distortion. Second, we employ EIT by subjecting it to the suspicious transition line, and try to eliminate it. As a result, the other transition line (the one that was hidden) can be seen. Third, we decipher the emerged transition line. We subject it to the right transition of the same or other molecule.

In our published paper [4], we derived the specific conditions that this method can be employed. Furthermore, we presented in this paper a few examples of the proposed method on real data. The data, given in the table below, consists of the overlapping transition frequencies of  $\text{Cl}_2$  isotopomers.

Isotopomers	$^{35,35}\text{Cl}_2$ $\nu_{2-9}$ band	$^{35,37}\text{Cl}_2$ $\nu_{1-9}$ band	$^{35,35}\text{Cl}_2$ $\nu_{1-9}$ band
Rotational branch $\omega_{\text{eg}} (\text{cm}^{-1})$	$R_{J=59}, 1.312 \times 10^4$	$P_{J=28}, 1.312 \times 10^4$	$P_{J=5}, 1.312 \times 10^4$
Rotational branch $\omega_{\text{eg}} (\text{cm}^{-1})$		$R_{J=32}, 1.3122 \times 10^4$	$R_{J=7}, 1.3122 \times 10^4$
Rotational branch $\omega_{\text{eg}} (\text{cm}^{-1})$	$P_{J=55}, 1.311 \times 10^4$	$R_{J=33}, 1.3119 \times 10^4$	

In summary, we analysed the conditions in which EIT phenomenon can increase tremendously the resolution of measuring (distinguishing) overlapping transition lines. This new model can also help in increasing the penetration depth of a probe beam due to the EIT, and can be exploited in the measurement of small minority of absorbers in mixtures of molecules.

v) Personnel Supported:

Dr. Evgeny Shapiro (Research Associate), Dr. Asaf Eilam (post-doctoral fellow), Dr. Gilad Hurvitz (Research Associate), Thomas Drane (graduate student) and Martin Bitter (graduate student).

vi) Publications:

1. Bitter M., Shapiro E.A. & Milner V. "Strong-field control of coherent anti-Stokes Raman scattering in iodine vapor with shaped ultrashort laser pulses", *To be submitted* (2012).
2. Drane T.M., Schlüter A., Milner A.A. & Milner V. "Polarization modulation spectroscopy with coherent anti-Stokes Raman scattering", *To be submitted* (2012).
3. Bitter M., Shapiro E. A., Milner V., Coherent ro-vibrational revivals in a thermal molecular ensemble. *Phys. Rev. A* 85, 043410 (2012).
4. A. Eilam, and M. Shapiro "Strong-field adiabatic passage in the continuum: Electromagnetically induced transparency and stimulated Raman adiabatic passage.," *Phys. Rev. A*, p. 012520, 2012.
5. A. Eilam, E. A. Shapiro, and M. Shapiro "Electromagnetically induced transparency spectroscopy," *J. Chem. Phys.*, vol. 136, p. 064201, 2012.
6. Shapiro E. A., Drane T. M. & Milner V. "Prospects of coherent control in turbid media: Bounds on focusing broadband laser pulses" *Phys. Rev. A* 84, 053807 (2011).
7. Zhdanovich S., Hepburn J. W. & Milner V. "Strong-field effects in Rabi oscillations between a single state and a superposition of states" *Phys. Rev. A* 84, 053428 (2011).
8. Shapiro, E. A., Konorov, S. O. & Milner, V. "Interference spectroscopy with coherent anti-Stokes Raman scattering of noisy broadband pulses" *J. Raman Spec.* (2011).
9. Konorov, S. O., Hepburn, J. W. & Milner, V. "Time- and frequency-resolved detection of atomic coherence in the regime of strong-field interaction with intense femtosecond laser pulses" *Phys. Rev. A* 83, 033417 (2011).
10. Drane T. M., Hepburn J. W. & Milner V. "Coherent Raman spectroscopy through one-dimensional random scattering medium" *J. Mod. Opt.* 57, 1928-1935 (2010).



11. Konorov S. O., Xu X. G., Hepburn J. W. & Milner V. "Characterization of transient molecular vibration excited with shaped femtosecond pulses" *J. Chem. Phys.* 130, 234505 (2009).

vii) Interactions/Transitions:

- a. Participation/presentations at meetings, conferences, seminars, etc.

Valery Milner

INVITED TALK

"Frequency resolved optical gating of atomic and molecular coherence: from weak to strong field regimes",

OSA's 95<sup>th</sup> Annual Meeting "Frontiers in Optics 2011"

San Jose, California,

October 16-20, 2011.

Evgeny Shapiro

INVITED TALK

"CARS spectroscopy by interfering noisy broadband light",

International conference "Physics of Quantum Electronics"

Snowbird, Utah,

January, 2011.

Evgeny Shapiro

"Driving molecules with pulse trains, and other developments in coherent control",

University of Rochester,

April, 2011.

Evgeny Shapiro

"Coherent nonlinear spectroscopy with noisy broadband laser pulses",

Harvard-Smithsonian Institute for Theoretical Atomic, Molecular and Optical Physics,

Harvard University,

April, 2011.

Evgeny Shapiro

"Engineering and Control of Quantum Systems",

Max Planck Institute for the Physics of Complex Systems,

Dresden, Germany,

October, 2011.

Evgeny Shapiro

"Piecewise adiabatic passage",

Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy,

Berlin, Germany,

October, 2011.

Valery Milner

INVITED TALK

"Narrow-band coherent spectroscopy with broad-band incoherent pulses"

40th Winter Colloquium on Physics of Quantum Electronics,

Snowbird, Utah,

January 4-8, 2010.

Valery Milner

INVITED TALK

"Nonlinear spectroscopy with coherently and incoherently shaped laser pulses"

239th American Chemical Society National Meeting

San Francisco, CA, March 21-25, 2010

viii) New discoveries, inventions, or patent disclosures. None

## **Final Report from JILA/University of Colorado - Boulder (Investigator: Ye)**

### **ii) Objectives:**

Development of frequency combs in spectral regions extending from mid infrared to extreme ultraviolet for direct frequency comb spectroscopy of complex molecular samples

### **iii) Status of effort:**

We have achieved the following milestones: (1) Performed the first ultrasensitive molecular spectroscopy in mid-infrared with frequency combs, combining FTIR with comb spectroscopy; (2) Developed a new technique that provides massively parallel comb spectroscopy sensitive specifically to ions through the combination of cavity-enhanced direct frequency comb spectroscopy with velocity-modulation spectroscopy; (3) Demonstrated a quantum-noise-limited absorption sensitivity with cavity-enhanced frequency comb spectroscopy, achieving the highest demonstrated sensitivity for a comb-based technique; (4) A groundbreaking work achieved during the funding period is the experimental demonstration of the existence of a frequency comb in the extreme ultraviolet (XUV) spectral region and performing the first spectroscopy work in the XUV spectral region.

### **iv) Accomplishments/New Findings:**

A groundbreaking work for the JILA group is the experimental demonstration of the existence of a frequency comb in the extreme ultraviolet region. A novel application of frequency comb technology that leverages both the ultrashort duration of each laser pulse and the exquisite phase coherence of a train of pulses is the generation of frequency combs in the extreme ultraviolet (XUV) via high harmonic generation (HHG) in a femtosecond enhancement cavity. Until now, this method has lacked sufficient average power for applications, which has also hampered efforts to observe phase coherence of the high-repetition rate pulse train produced in the extremely nonlinear HHG process. Hence, the existence of a frequency comb in the XUV has not been confirmed. The group has overcome both challenges and has achieved generation of  $>200\text{ }\mu\text{W}$  per harmonic reaching 50 nm ( $20\text{ }\mu\text{W}$  after harmonic separation). In a true groundbreaking fashion, the group made the observation of single-photon spectroscopy signals for both an argon transition at 82 nm and a neon transition at 63 nm. The absolute frequency of the argon transition has been determined via direct frequency comb spectroscopy. The resolved 10-MHz linewidth of the transition, limited by the transverse temperature of the argon atoms, is unprecedented in this spectral region and places a stringent upper limit on the linewidth of individual comb teeth. Due to the lack of continuous wave lasers, these frequency combs are currently the only promising avenue towards extending ultrahigh precision spectroscopy to below the 100-nm spectral region with a wide range of applications that include spectroscopy of

electronic transitions in molecules, experimental tests of bound state and many body quantum electrodynamics in  $\text{He}^+$  and  $\text{He}$ , development of next-generation “nuclear” clocks, and searches for spatial and temporal variation of fundamental constants using the enhanced sensitivity of highly charged ions.

The JILA group has also successfully implemented the first high-resolution frequency comb spectroscopy in the mid infrared region. Optical frequency comb-based high-resolution spectrometers offer enormous potential for spectroscopic applications. Although a variety of implementations have been demonstrated, the lack of suitable mid-infrared comb sources has impeded explorations of molecular fingerprinting. The group for the first time implemented a frequency-comb Fourier transform spectrometer operating in the 2100-to-3700  $\text{cm}^{-1}$  spectral region that allows fast and simultaneous acquisitions of broadband absorption spectra with up to 0.0056  $\text{cm}^{-1}$  resolution. They demonstrated part-per-billion detection limits in 30 seconds of integration time for various important molecules including methane, ethane, isoprene, and nitrous oxide. The system enables precise concentration measurements even in gas mixtures that exhibit continuous absorption bands, and it allows detection of molecules at levels below the noise floor via simultaneous analysis of multiple spectral features. This system represents a near real-time, high-resolution and high-bandwidth mid-infrared spectrometer which is ready to replace the traditional Fourier transform spectrometers for many applications in trace gas detection, atmospheric science, and medical diagnostics. Recently the JILA group has also demonstrated a quantum-noise-limited absorption sensitivity with cavity-enhanced frequency comb spectroscopy, achieving the highest demonstrated sensitivity for a comb-based technique. The system comprises a frequency comb locked to a high-finesse cavity and a fast-scanning Fourier transform spectrometer with an ultra-low-noise autobalancing detector. Spectra with a signal-to-noise ratio above 1000 and a resolution of 380 MHz are acquired within a few seconds. The measured absorption lineshapes are in excellent agreement with theoretical predictions. Finally, in collaboration with Eric Cornell’s group, they have demonstrated a new technique that provides massively parallel comb spectroscopy sensitive specifically to ions through the combination of cavity-enhanced direct frequency comb spectroscopy with velocity-modulation spectroscopy..

v) Personnel Supported:

Dr. Aleksandra Foltynowicz and Dr. Piotr Masłowski (Research Associates, postdocs), Kevin Cossel and Bryce Bjork (Research Assistant, graduate student).

vi) Publications:

1. F. Adler, P. Masłowski, A. Foltynowicz, K. C. Cossel, T. C. Briles, I. Hartl, and J. Ye, “Mid-infrared Fourier transform spectroscopy with a broadband frequency comb,” *Opt. Express* **18**, 21861 – 21872 (2010).
2. A. Foltynowicz, P. Masłowski, T. Ban, F. Adler, K. C. Cossel, T. C. Briles, and J. Ye, “Optical frequency comb spectroscopy,” *Faraday Disc.* **150**, 23 – 31 (2011).
3. A. Ruehl, M. J. Martin, K. C. Cossel, L. Chen, H. McKay, B. Thomas, L. Dong, C. Benko, J. M. Dudley, M. E. Fermann, I. Hartl, and J. Ye, “Ultra-broadband coherent frequency comb,” *Phys. Rev. A* **84**, 011806, Rapid Communications (2011).
4. L. Sinclair, K. C. Cossel, T. Coffey, J. Ye, and E. Cornell, “Frequency comb velocity-modulation spectroscopy,” *Phys. Rev. Lett.* **107**, 093002 (2011).
5. T. K. Allison, A. Cingöz, D. C. Yost, and J. Ye, “Extreme nonlinear optics in a femtosecond enhancement cavity,” *Phys. Rev. Lett.* **107**, 183903 (2011).
6. A. Foltynowicz, T. Ban, P. Masłowski, F. Adler, and J. Ye, “Quantum noise limited optical frequency comb spectroscopy,” *Phys. Rev. Lett.* **107**, 233002 (2011). (Editor’s Suggestion; Selected for a Viewpoint in *Physics*.)
7. D. C. Yost, A. Cingöz, T. K. Allison, A. Ruehl, M. E. Fermann, I. Hartl, and J. Ye, “Power optimization of XUV Frequency Combs for Spectroscopy Applications,” *Opt. Express* **19**, 23483 – 23493 (2011). (Invited Article)
8. A. Cingöz, D. C. Yost, T. K. Allison, A. Ruehl, M. E. Fermann, I. Hartl, and J. Ye, “Direct frequency comb spectroscopy in the extreme ultraviolet,” *Nature* **482**, 68 – 71 (2012).

vii) Interactions/Transitions:

- a. Participation/presentations at meetings, conferences, colloquia and seminars, etc.

The following talks are all invited presentations.

1. D. Felinto and J. Ye, “Direct frequency comb spectroscopy,” Latin America Optics and Photonics Conference (LAOP), Recife, Brazil, September 27 – 30, 2010.
2. T. Allison, D. C. Yost, A. Cingöz, A. Ruel, and M. E. Fermann, I. Hartl, and J. Ye, “The Extreme Ultraviolet Frequency Comb and Prospects for X-ray Combs,” Workshop on Evolution and Control of Complexity: Key Experiments Using Sources of Hard X-rays, The Advanced Photon Source/Argonne National Laboratory, October 11 – 13, 2010.
3. A. Ruehl, M. E. Fermann, I. Hartl, A. Cingöz, D. C. Yost, and J. Ye, “High Power Fiber Laser Frequency Combs for XUV Spectroscopy,” Optical Society of America, Fiber Laser Applications (FILAS), Istanbul, Turkey, February 16 – 17, 2011.

4. J. Ye, "New opportunities for precision spectroscopy," Faraday Discussions on "Frontiers in Spectroscopy" University of Basel, Switzerland, April 6 – 8, 2011.
5. J. Ye, "Frequency comb spectroscopy – from IR to XUV," OSA/SPIE Student Chapter, Stanford University, Stanford, California, May 5, 2011.
6. P. Masłowski, A. Foltynowicz, F. Adler, K. C. Cossel, T. C. Briles, T. Ban, and J. Ye, "Mid-infrared frequency comb spectroscopy," Conference on Lasers and Electro-Optics (CLEO), Baltimore, Maryland, May 2 – 6, 2011.
7. A. Foltynowicz and J. Ye, "Cavity-enhanced optical frequency comb spectroscopy," Cavity Enhanced Spectroscopy Conference, Kingston, Canada, June 26 – 29, 2011. (Plenary)
8. J. Ye, "Frequency combs and spectroscopy - from IR to XUV," Extreme Photonics Summer School, Ottawa, Canada, June 24 – 30, 2011.
9. T. Ban, A. Foltynowicz, P. Masłowski, D. Aumiler, G. Pichler, and J. Ye, "Direct frequency comb spectroscopy: time and frequency domain approach," The Forty Third Conference of the European Group on Atomic Systems (EGAS-43), Fribourg, Switzerland, June 28 – July 2, 2011.
10. J. Ye, "Frequency combs from mid-IR to XUV," OSA Topical Meeting on Nonlinear Optics, Kaua'i Marriott resort, Lihue-Kauai, Hawaii, July 17 – 22, 2011.
11. J. Ye, "Coherent control for cold molecules," Gordon Research Conference on Quantum Control of Light and Matter, Mt. Holyoke College, Massachusetts, Jul. 31 – Aug. 5, 2011.
12. J. Ye, "Frequency combs and spectroscopy - from IR to XUV," OSA Traveling Lecture, OSA Student Chapter of Griffith University, Brisbane, Australia, August 22, 2011.
13. A. Foltynowicz-Matyba and J. Ye, "Frequency comb spectroscopy and applications," Field Laser Applications in Industry and Research (FLAIR) Conference, Murnau Germany, September 13 – 17, 2011.
14. K. C. Cossel, A. Foltynowicz, P. Masłowski, T. Ban, F. Adler, and J. Ye, "Mid-infrared and cavity-enhanced FTIR frequency comb spectroscopy," Symposium on *A Revolution in Spectroscopy by the Optical Frequency Combs*, Tsukuba, Japan, September 26, 2011.
15. J. Ye, "Precision measurements in many-particle systems," Annual Meeting, the Four Corners Section, American Physical Society, Tucson, Arizona, October 21 – 22, 2011.

16. J. Ye, "Frequency comb spectroscopy - from IR to extreme ultraviolet," Joint Seminar Series at Lund Laser Centre (LLC), "Quantum metrology with ultracold atoms - Optical atomic clocks," Physics Seminar, Lund University, Sweden, November 14 – 15, 2011.
17. J. Ye, "Direct frequency comb spectroscopy in the XUV," Atomic and Molecular Physics Program, AFOSR, Arlington, VA, January 5 – 6, 2012.
18. J. Ye, "Ultracold physics and ultrafast technology," Inaugural Winter School on Atomic, Molecular and Optical Physics, B2 Institute, Biosphere 2 Campus, University of Arizona, Tucson, Arizona, January 8 – 13, 2012. (Organized by: ITAMP & B2 Institute)
19. F. Adler, P. Maslowski, A. Foltynowicz, T. Ban, K. C. Cossel, and J. Ye, "Frequency comb spectroscopy in the mid-infrared for gas analysis," PITTCON, Specialty Gas Analysis Session, Philadelphia, PA, March 17 – 21, 2012.
20. J. Ye, "Probing and controlling quantum matter with precision light," Department of Physics, Columbia University, New York, March 29, 2012.
21. J. Ye, "Frequency comb spectroscopy from mid-infrared to extreme ultraviolet," Conference Tutorial, Conference on Lasers and Electro-Optics (CLEO), San Jose, California, May 6-11, 2012.

viii) New discoveries, inventions, or patent disclosures. None



# Manipulation of molecular quantum wavepackets with ultrashort laser pulses for non-destructive detection of volatile explosives.

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We investigated the effect of quantum accumulation of laser-induced coherence in gaseous media for the purpose of selective excitation and detection of quantum ro-vibrational molecular wavepackets with frequency comb sources, and with trains of ultrashort laser pulses.

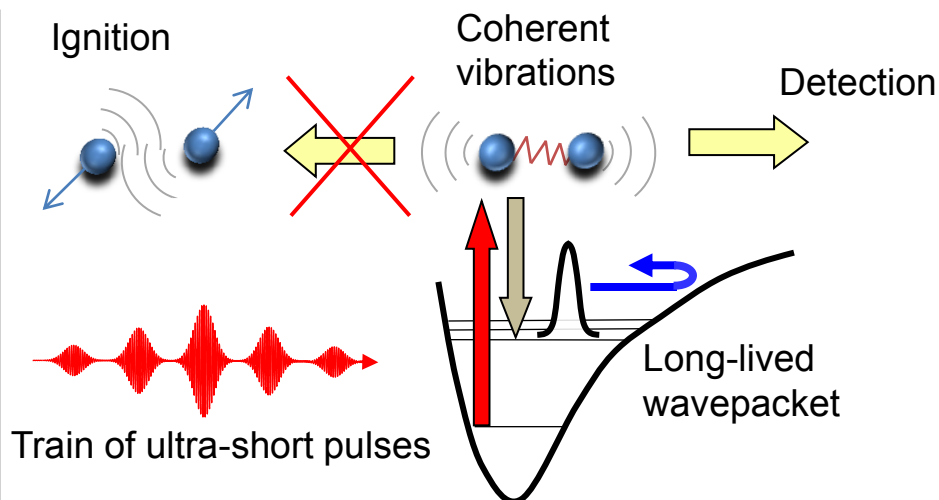
## Challenges addressed:

- enhance the sensitivity to the level suitable for standoff detection, despite low molecular concentration;
- prevent undesired heating of gas samples despite the required high laser power.
- withstand de-coherence

Status of effort: completed theoretical and experimental work on six methods of molecular detection with shaped femtosecond pulses; performed ultrasensitive molecular spectroscopy with mid-infrared frequency combs, the existence of a frequency comb in the extreme ultraviolet (XUV) spectral region demonstrated.

Personnel supported: At UBC: 3 post-doctoral fellow, 2 graduate students; at JILA/UC-Boulder: 2 post-doctoral fellow, 1 graduate student.

Publication & Meetings: 19 papers published, 29 invited and contributed talks at conferences and symposia.



- Year 1: development of a theoretical model; design and implementation of two experimental setups;
- Year 2: experiments in room-temperature and cold simple molecular systems; theoretical analysis;
- Year 3: experiments with complex molecules related to volatile explosives; analysis of de-coherence.

Year 1 (Jan 09 – Dec 09)	Year 2 (Jan 10 – Dec 10)	Year 3 (Jan 11 – Dec 11)
\$ 407,401	\$358,974	\$359,652

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