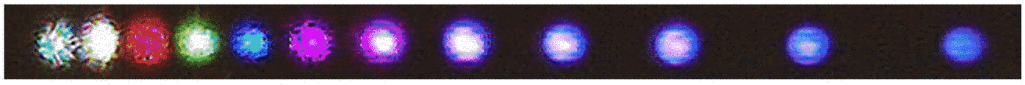
KITP workshop on Attosecond Science
Santa Barbara, August 1 – 4, 2006

Broadband coherent Raman generation in gasses and solids



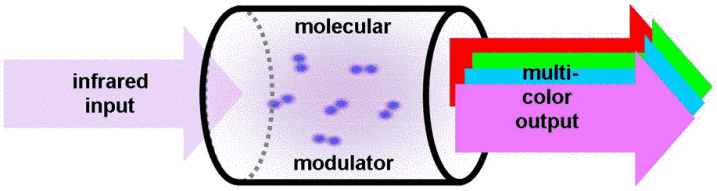
Alexei Sokolov, Andrea Burzo, Alexey Chugreev,
Miaochan Zhi, Xi Wang, Jiahui Peng, Dmitry Pestov

Physics Department and Institute for Quantum Studies
Texas A&M University

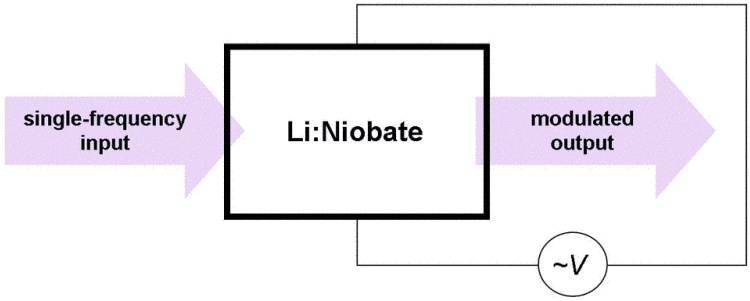
Molecular Modulation leads to generation of broad coherent bandwidth and allows production of ultrashort pulses, opening a range of exciting possibilities:

- sub-cycle field shaping and non-sinusoidal wave generation;
- synchronization of light pulses with molecular oscillations, with a possibility of tomographic imaging of vibrating molecules;
- possible application to controlled nuclear collisions.

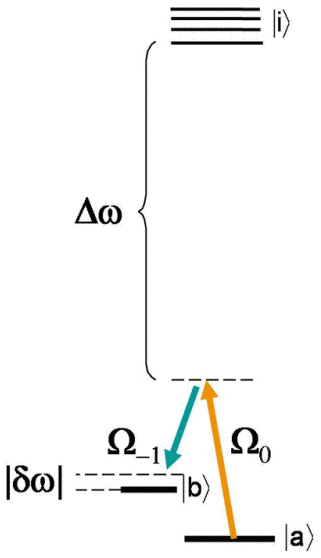
Molecular Modulation:



Is in essence similar to electro-optical modulation:



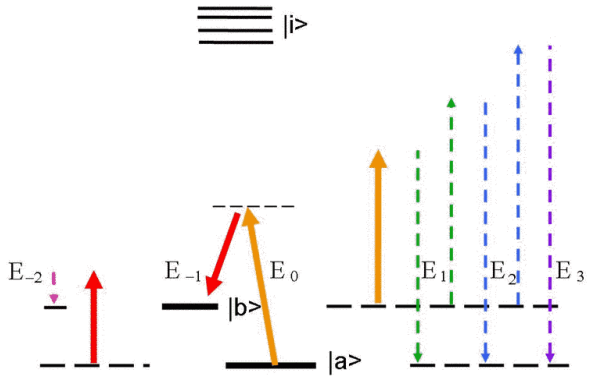
Strongly driven molecular systems



$$\frac{\Omega_0\,\Omega_{-1}}{\Delta\omega} > |\delta\omega|$$

$$|\rho_{ab}| = \rho_{aa} = \rho_{bb} = \frac{1}{2}$$

Multiple sideband generation



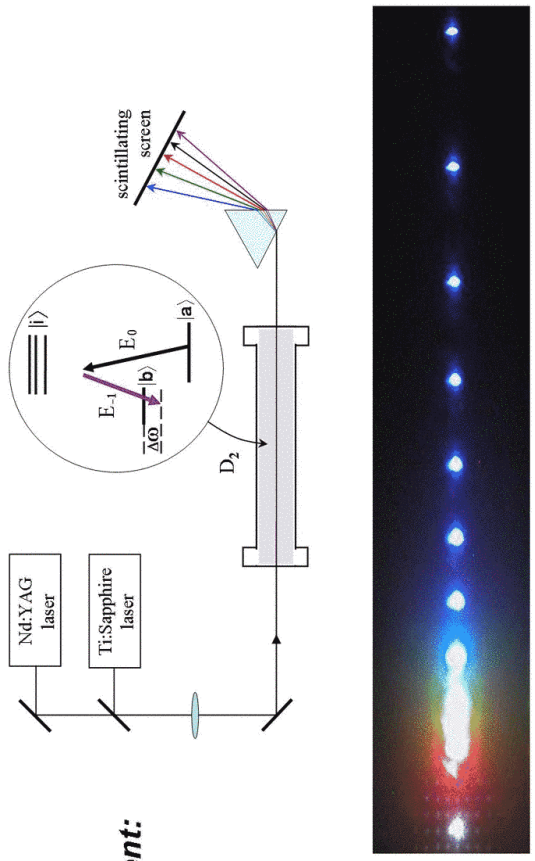
$$\frac{\partial E_q}{\partial z} = -j\eta\hbar\omega_q N\left(a_q\rho_{aa}E_q + d_q\rho_{bb}E_q + b_q^*\rho_{ab}E_{q-1} + c_q\rho_{ab}^*E_{q+1}\right)$$

Rough outline:

(1) Molecular modulation in gasses, in nanosecond regime.

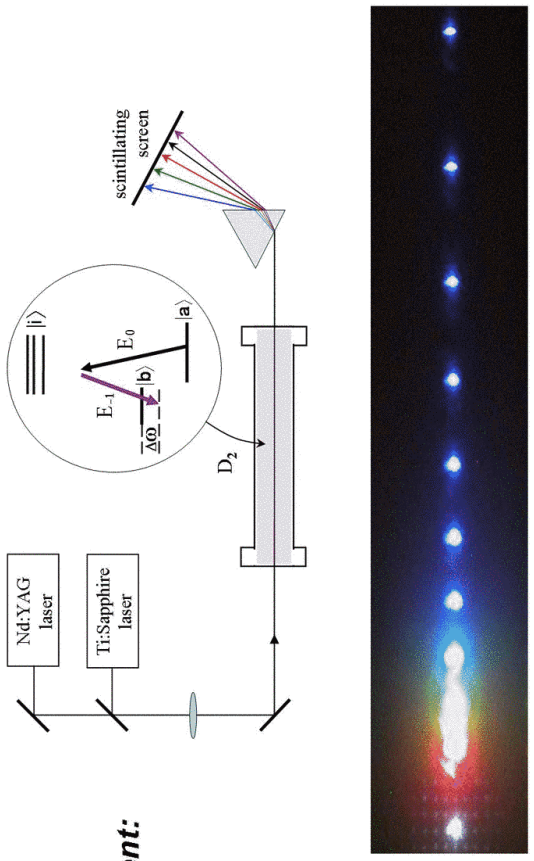
(2) Efficient broad-band generation in Raman-active crystals driven by femtosecond pulses.

In both regimes the driving pulse duration is comparable to the coherence lifetime.



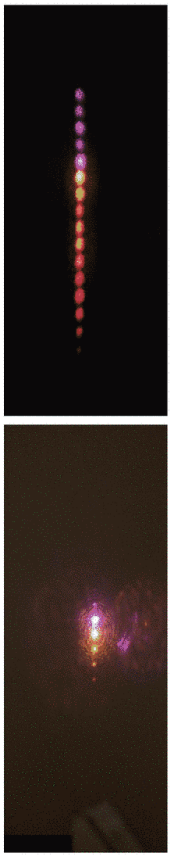
Experiment:

vibrational Raman generation



Experiment:

vibrational Raman generation

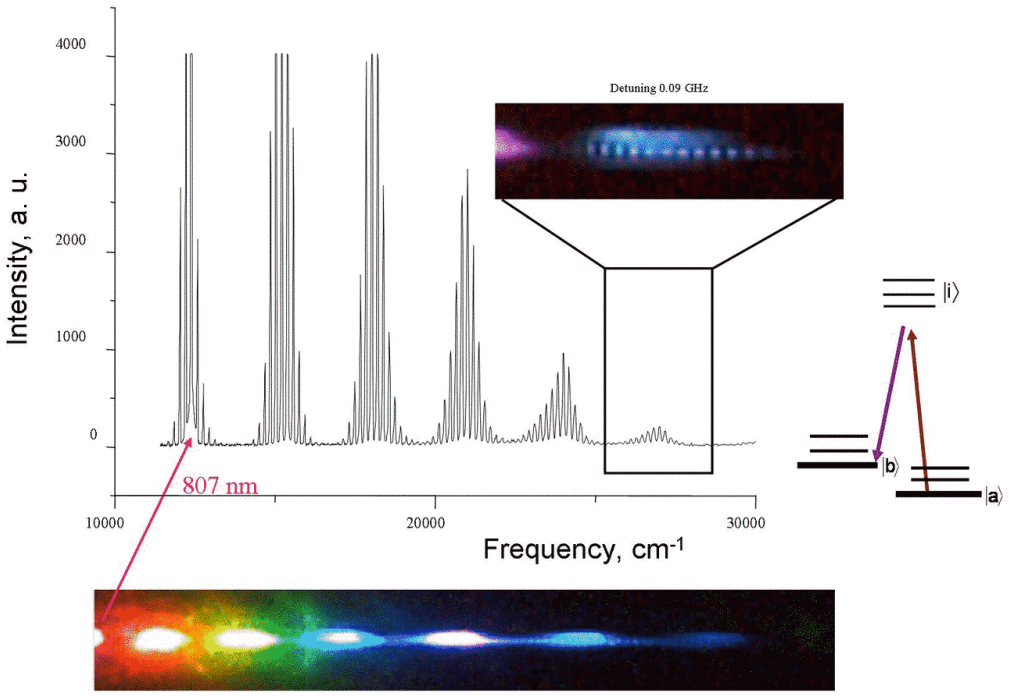


rotational Raman generation

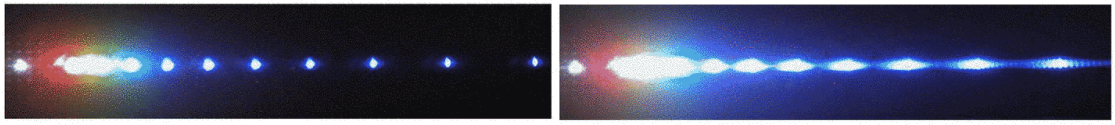
in free space

in a hollow waveguide

Simultaneous vibrational and rotational generation



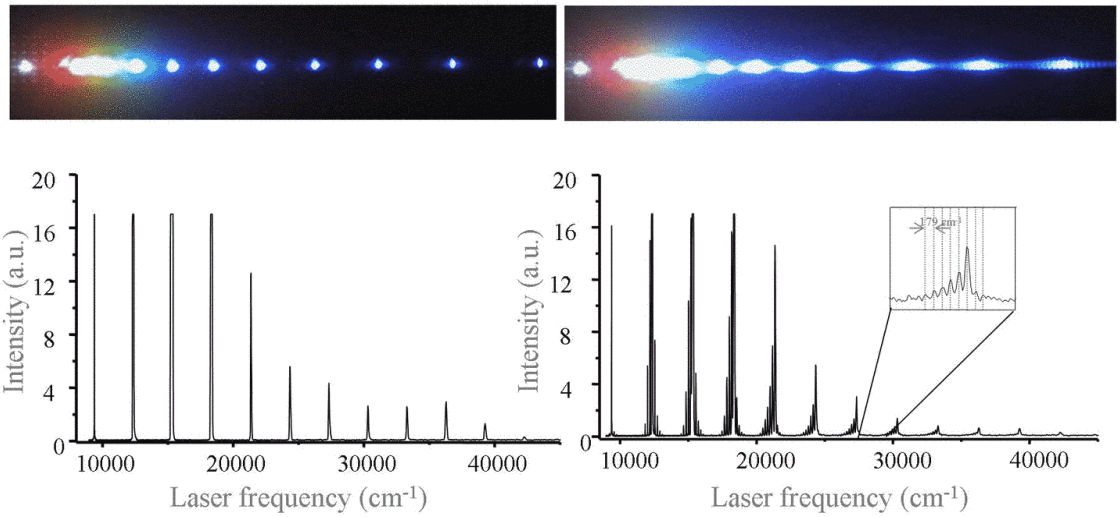
Simultaneous vibrational and rotational generation at different Raman detunings



vibrational only

vibrational and rotational

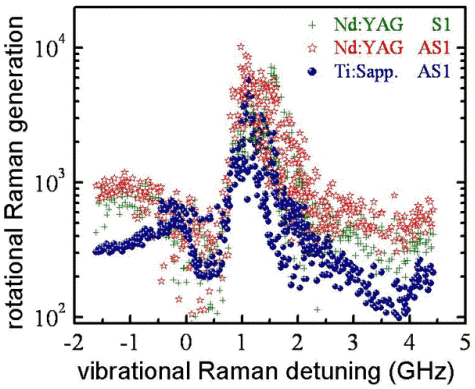
Simultaneous vibrational and rotational generation at different Raman detunings



vibrational only

vibrational and rotational

Stimulated rotational Raman scattering is enhanced by the vibrational generation



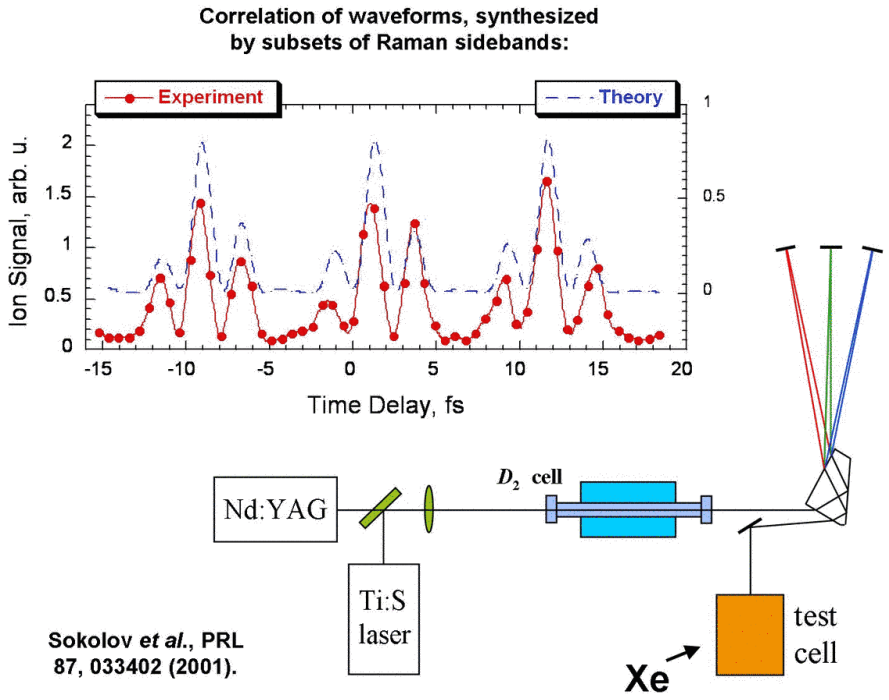
We are trying to sort out what is going on. Some of the possibilities are:

- Raman selffocusing (enhanced by the efficient vibrational generation);

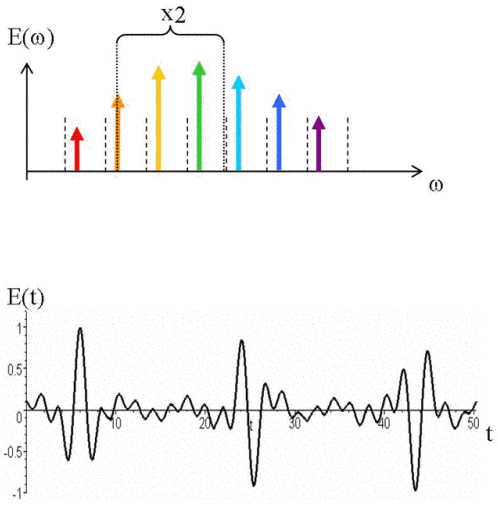
- ultrashort pulse compression by dispersion of the molecular medium;

- EIT-like quantum interference within the molecular system.

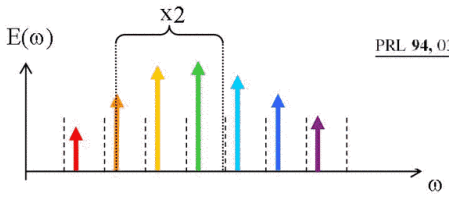
We have used multiphoton ionization of xenon in order to characterize the synthesized pulse train.



Sub-cycle phase selection



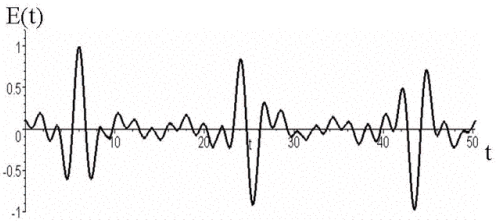
Sub-cycle phase selection

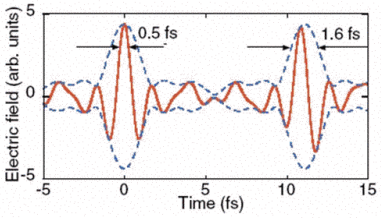


PRL 94, 033904 (2005) PHYSICAL REVIEW LETTERS

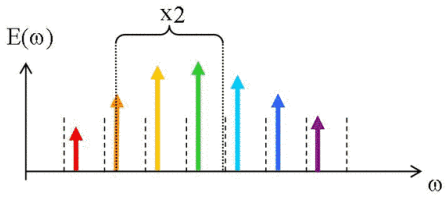
Generation of a Single-Cycle Optical Pulse

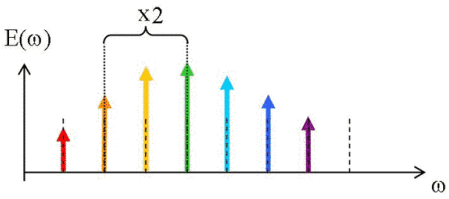
M. Y. Shverdin, D. R. Walker, D. D. Yavuz,* G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA

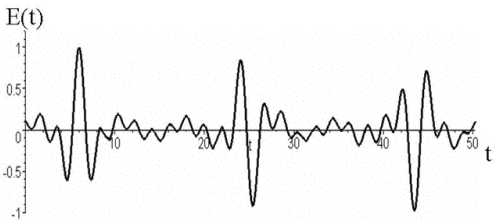




Sub-cycle phase selection

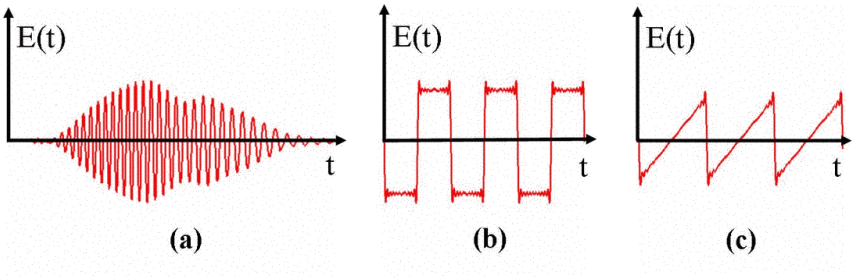




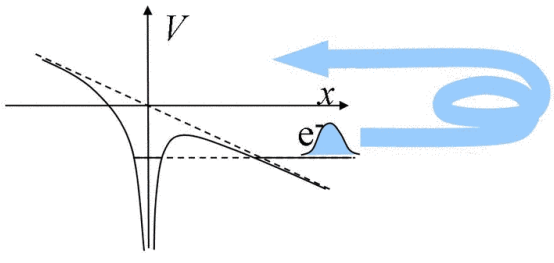


Pulse shaping: "old" and "new"

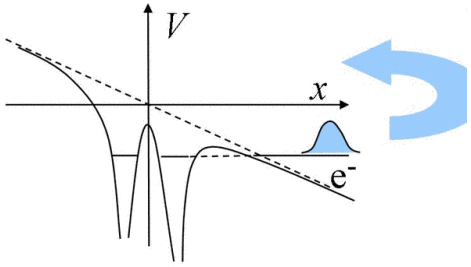
(a) "Traditional" pulse shaping; (b) and (c) Sub-cycle pulse shaping.



Atom (or molecule) in a strong sub-cycle-shaped laser field.

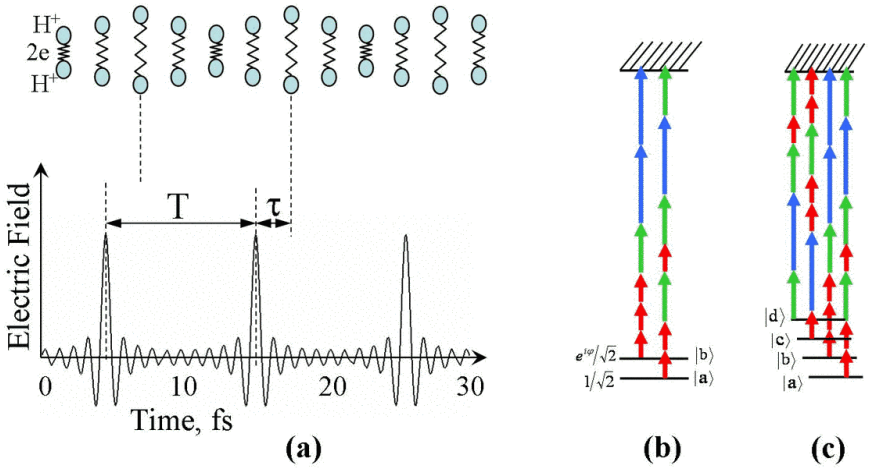


The electron trajectory,
the recollision time,
and the recollision energy
can be controlled precisely.

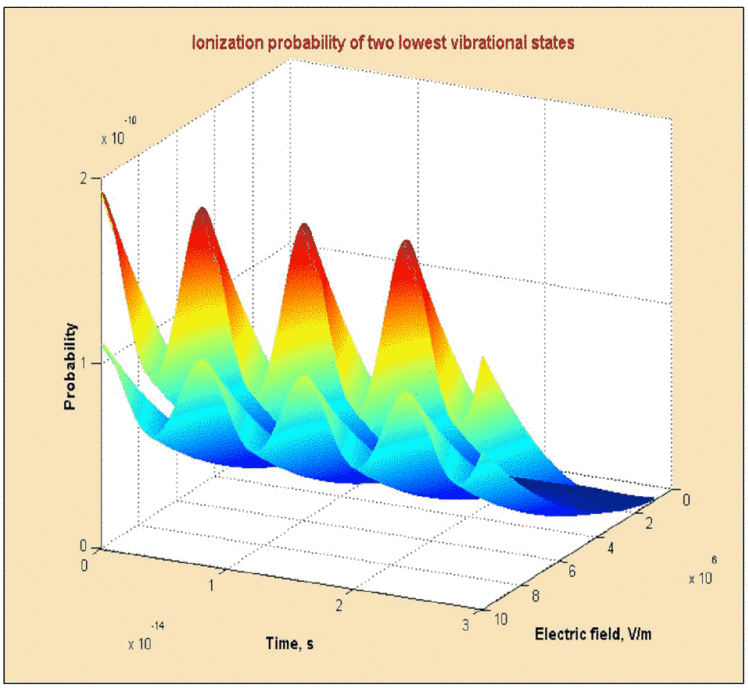


HHG in molecules:
the electron "scans"
the spatial molecular
structure.

Molecular oscillations modulate light and produce trains of pulses, which are (by the very nature of the modulation process) perfectly synchronized with the molecular motion.



Ionization probability as a function of time delay between a train of single-cycle (sin) pulses and the phase of the molecular motion which corresponds to a coherent superposition of the ground and first excited vibrational states of H_2+



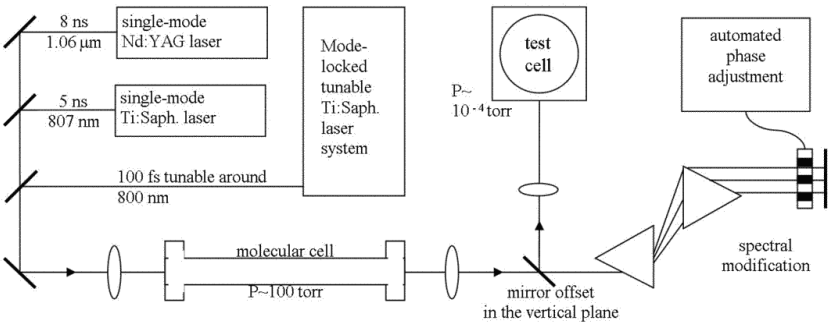
Calculation by A. Burzo using the Keldysh model

Can single pulses be obtained by MM?

Probably YES,

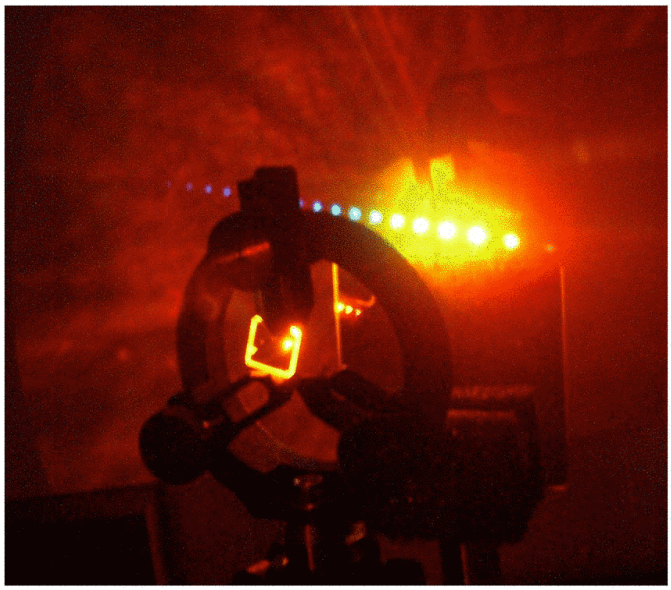
by applying a femtosecond pulse
to the molecular ensemble
driven by the two nanosecond pulses.

Schematics of the proposed experimental setup, which will combine adiabatic preparation of the maximal Raman coherence by two nanosecond lasers, a femtosecond probe laser (whose pulses will be compressed into the sub-femtosecond domain), and a spectral modification setup (which will produce a desired sub-cycle pulse shapes in the test cell).

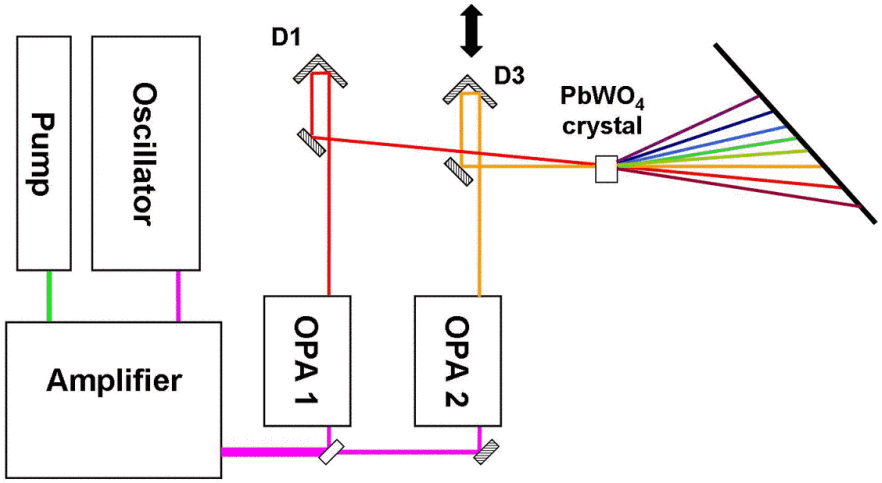


Molecular coherence can be excited (and probed) by femtosecond (or picosecond) pulses. Moreover, it can be done in a solid!

$PbWO_4$ (led tungstate) is a recently invented Raman crystal, with several strong and narrow Raman lines. However, the primary motivation for its development and mass-production has been its use as a scintillator in high-energy physics experiments.

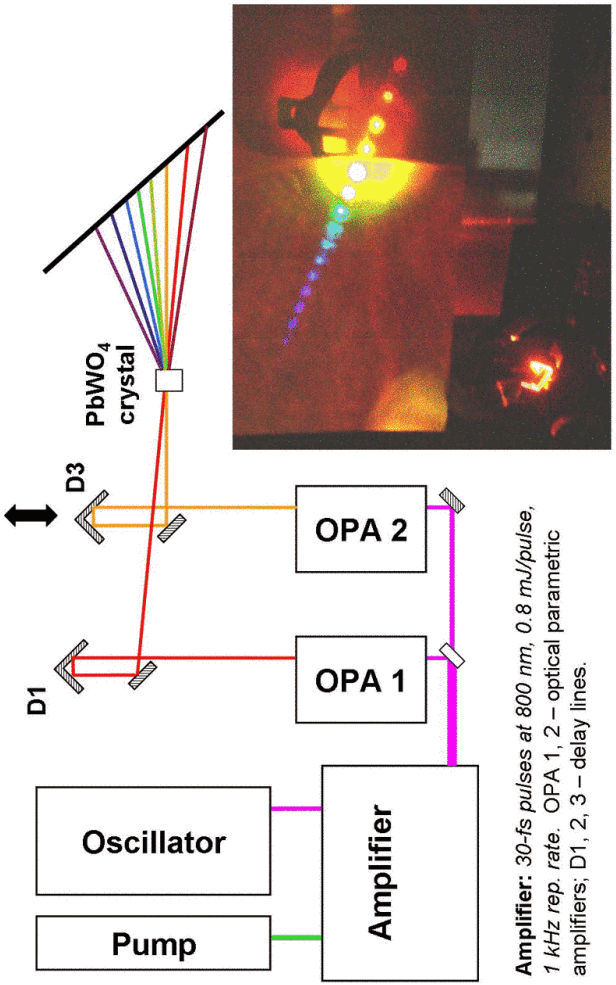


Femtosecond experiments with a Raman crystal



Amplifier: *30-fs pulses at 800 nm, 0.8 mJ/pulse, 1 kHz rep. rate.* OPA 1, 2 – optical parametric amplifiers; D1, 2, 3 – delay lines.

Femtosecond experiments with a Raman crystal



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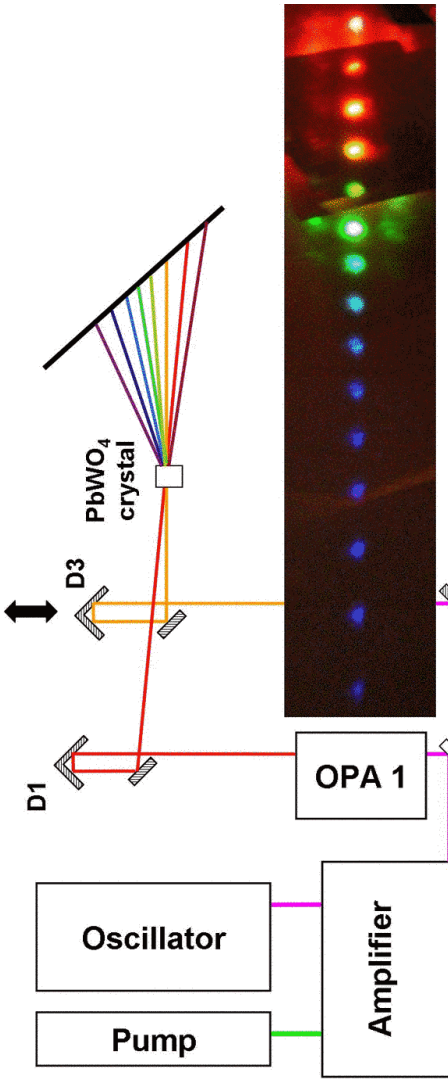
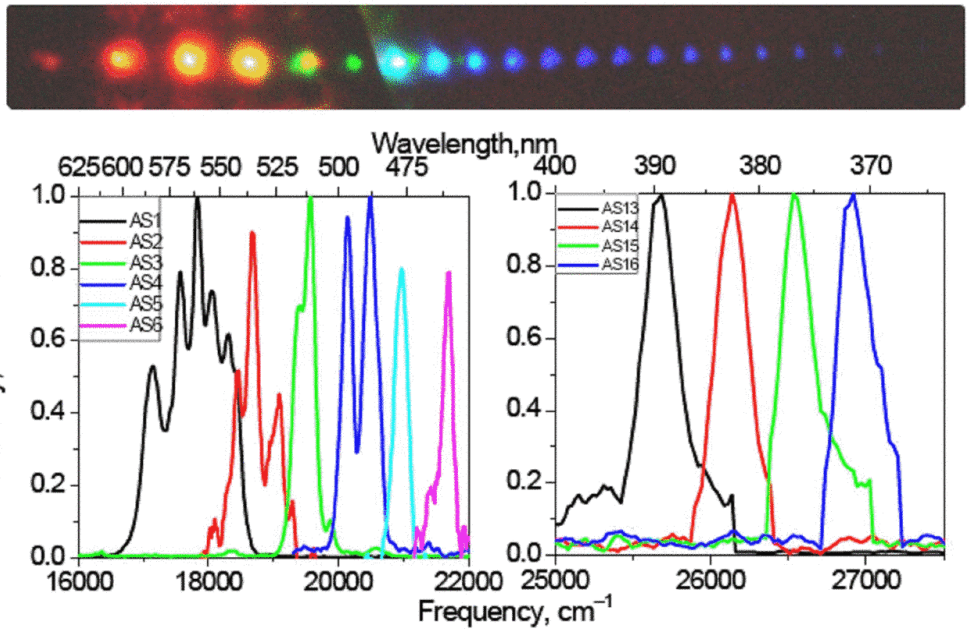


Broadband Raman generation in $PbWO_4$ (led tungstate)

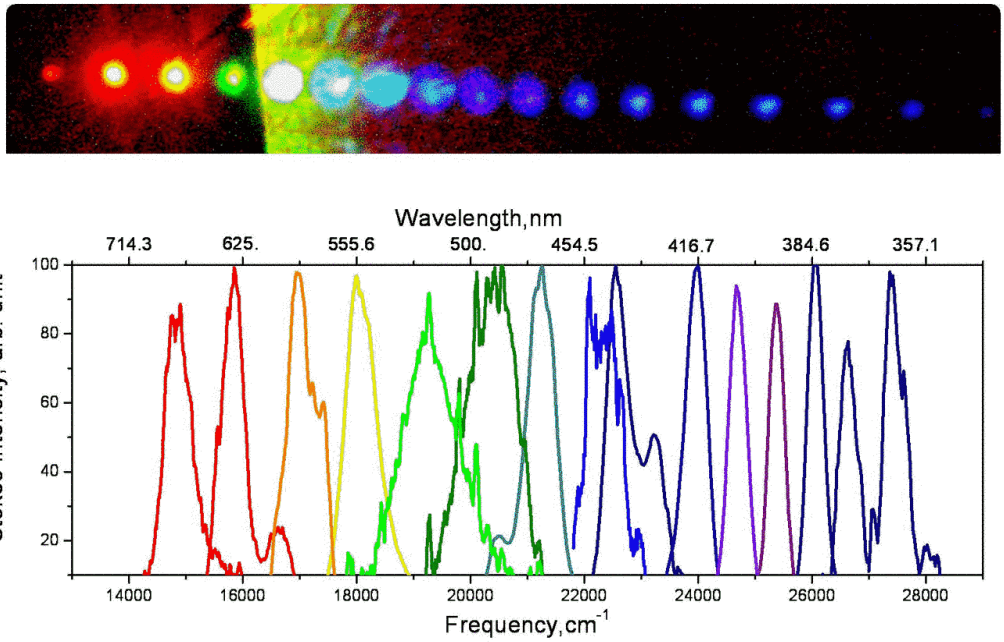
Amplifier: *30-fs pulses at 800 nm, 0.8 mJ/pulse, 1 kHz rep. rate.* OPA 1, 2 – optical parametric amplifiers; D1, 2, 3 – delay lines.

Generated spectrum consists of several (infrared) Stokes and up to 20 anti-Stokes sidebands that cover visible and UV spectrum.



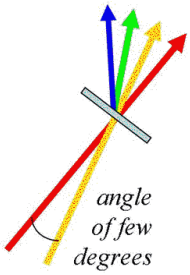
We measure the spectrum of individual sidebands by a fiber-coupled spectrometer.

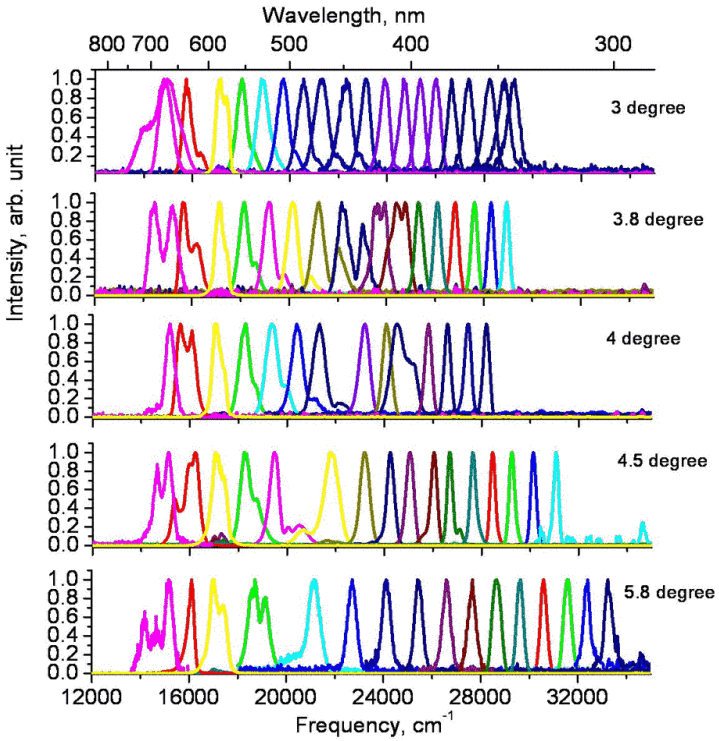
We observe similar broadband Raman generation in Diamond



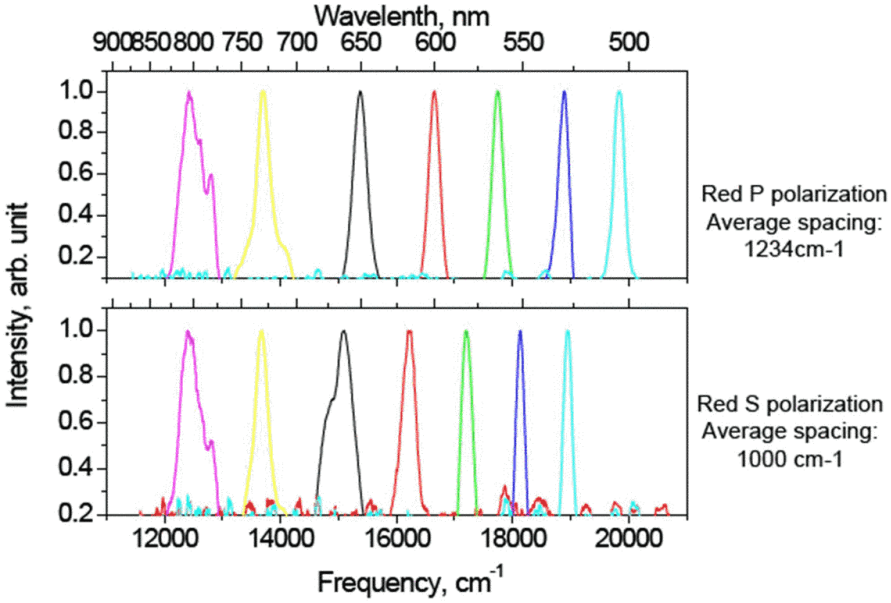
Apparently, phasematching plays a critical role in the generation of spectral sidebands.

As we vary the angle between the applied beams, the generated spectrum changes.



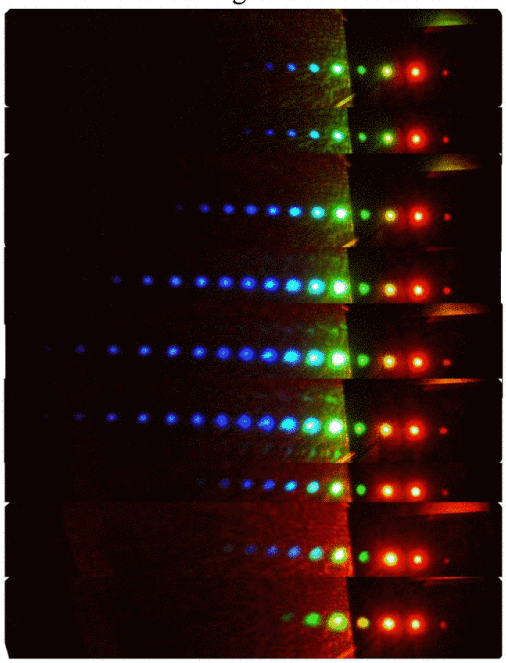


When we simply change the polarization of one of the applied beams, the frequency spacing of the generated spectral comb changes.



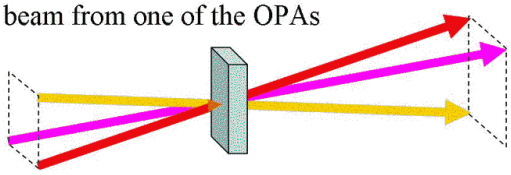
When we tune the difference frequency between the applied pump and Stokes fields, the generated spectrum is strongly affected.

Broadband Raman generation in Diamond

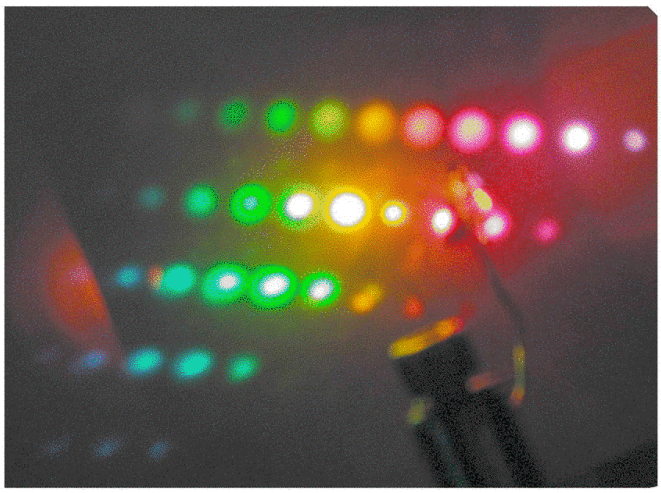


Stokes wavelength	pump wavelength is fixed at	difference frequency
572nm	631nm	1635 cm-1
577nm		1483cm-1
582nm		1334cm-1
587nm		1188cm-1
592nm		1043cm-1
597nm		903cm-1
602nm		763cm-1
607nm		627cm-1
612nm		492cm-1

add another (third) beam from one of the OPAs

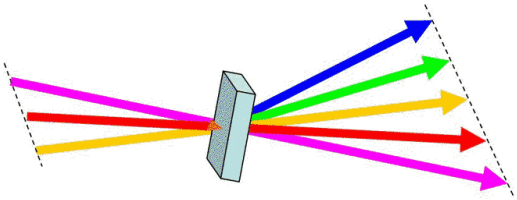


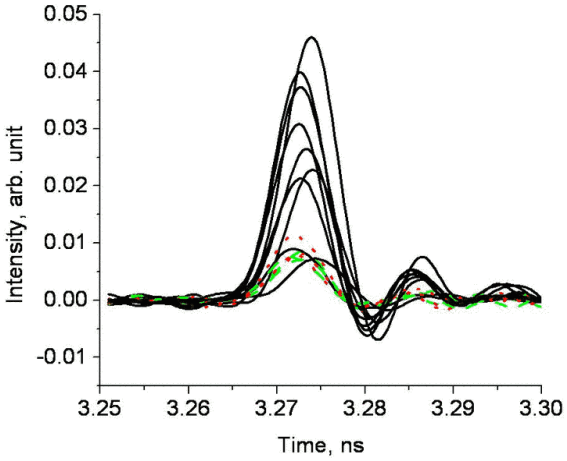
When we add a third beam at the input of the Raman crystal, we observe a 2D pattern of generated beams at the output.



When we align the three input beams in one plane, such that the higher-order anti-Stokes sidebands generated through different channels overlap in space, we observe their interference.

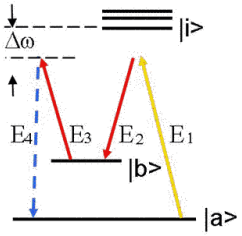
This is a signature of the sidebands' mutual coherence.

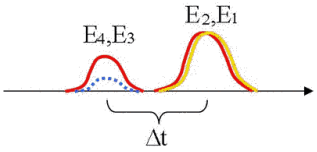




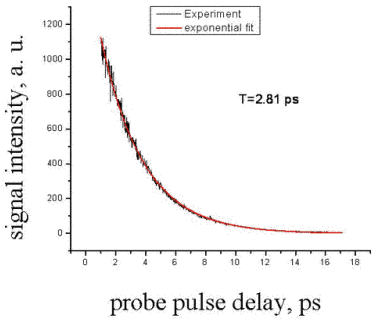
Black solid line: all three pulses present;
Red dotted line: two pulses present (Yellow blocked);
Green dashed line: two pulses present (IR blocked).

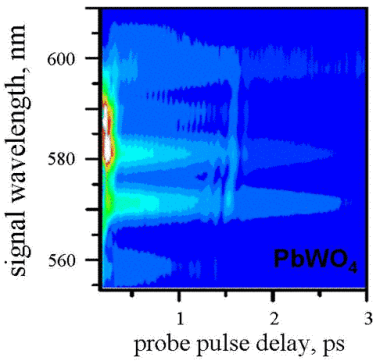
Femtosecond pump-probe experiments show exponential decay of Raman coherence with a characteristic time of a picosecond.

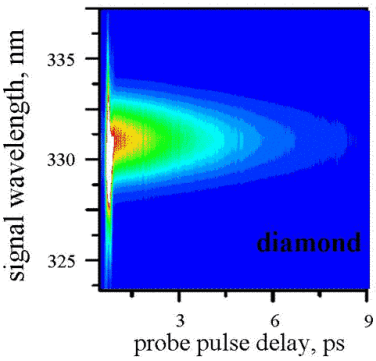




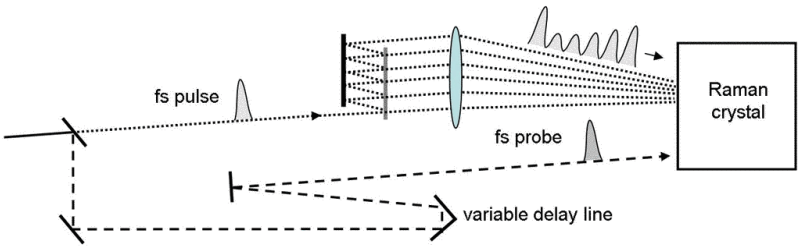
Typically, the pump and Stokes wavelength are in Red and Near-IR







Excitation of molecular coherence in the Raman crystal will be optimized if slightly longer (picosecond) pulses are used. A pair of such pulses can come from two separate OPA beams. Another possibility: *use an amplitude-modulated femtosecond waveform produced from a femtosecond pulse, in order to properly excite the molecular oscillation.*



Conclusions:

- Molecular modulation in nanosecond regime produces well-controlled sets of Raman sidebands, possibly resulting in generation of precisely shaped subfemtosecond pulses.
- Efficient broad-band molecular modulation is demonstrated in a Raman crystal driven by femtosecond pulses, opening exciting possibilities for the production of single femtosecond pulses.

Our Subfemtosecond group:

- Jiahui Peng
- Miaochan Zhi
- Dmitry Pestov
- Alexei Sokolov
- Alexey Chugreev
- Andrea Burzo

Other members of the FAST CARS team:

Xi Wang,
Robert Murawski,
Nikolai Kalugin,
Vladimir Sautenkov,
and Marlan Scully

$$$: NSF, The Welch Foundation, Research Corporation, DARPA