Santa Barbara, 2002

Slow Light in Solid Hydrogen

. . . . . .

Kohzo Hakuta

University of Electro-Communications
Chofu, Tokyo 182-8585, Japan

Outline of the Talk

Why Solid Hydrogen?

What is Solid Hydrogen?

How to prepare Solid Hydrogen crystal.

How good is it?
High-Resolution Raman Spectroscopy

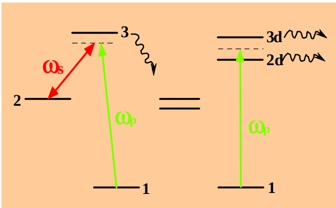
Nonlinear Optics in Solid Hydrogen
Arbitrary & Efficient Parametric Raman Sideband Generation

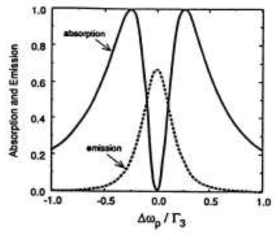
• *Slow Light in a Transparent Medium*

Possible Resonant EIT System in Solid Hydrogen

Background

- Nonlinear Optical Processes.
  Interplay between linear and nonlinear susceptibilities
  Linear Response . . Minimize . Nonlinear Response . . Maximize
  . . . . . . **Trade.off**
- Nonlinear Optics Using.Strong-Field Coupling and Induced Transparency. . (Electromagnetically.Induced.Transparency : EIT)





- Demonstration.H-atom (Hakuta, Stoicheff), Pb-atom (Harris Group at Stanford)
- Atomic System.*Slow Dephasing, Nearly Resonant*
- *Is it possible to extend to the "Condensed Phases".*
- *If O.K* Far Off-Resonant System
- Near Resonance -> New Manipulation Freedom

How to realize 'High Density" and "Narrow Raman Width", Simultaneously ?

Solid Hydrogen – Quantum Crystal
Molecular Crystal with Quantized Vibrational-Rotational Motion

 # Vibrational Quantum number $\upsilon = \sum_{i=1}^{N} \upsilon_i$

 # Ground Vibrational State; v=0 $\langle x_1 \cdots x_N | 0 \rangle = \prod_{i=1}^{N} \phi_0(x_i)$

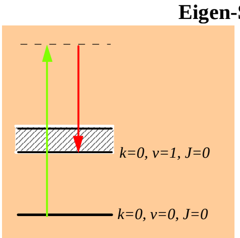
"No Degeneracy"

 # First Excited State ; v=1 $\langle x_1 \cdots x_N | R_i \rangle = \phi_1(x_i) \prod_{j \neq i} \phi_0(x_i)$

"N-Fold Degeneracy"

Hopping Interaction Between Sites

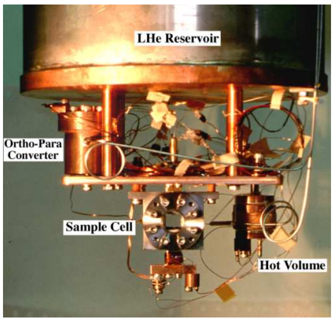
Eigen-State $| \rangle = N^{-1/2} \sum_{i=1}^{N} \exp(ik \cdot R_i) |R_i\rangle$

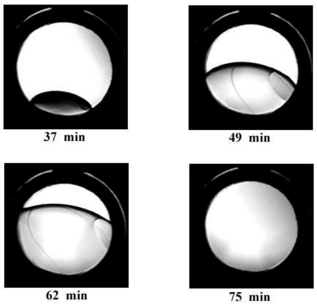


Raman tansition from the ground state

Ground State Vibron with k=0

Crystal growth from Liquid Phase





Para-Hydrogen > 99.9 %

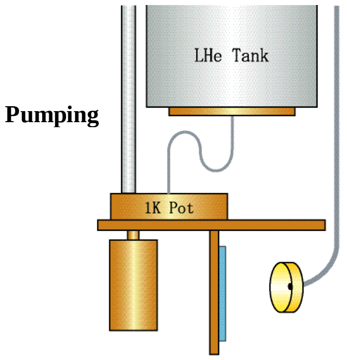
Cell Length: 1 cm

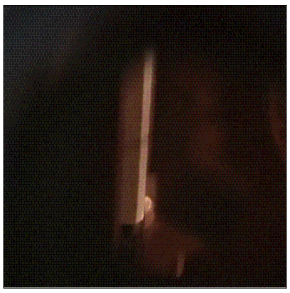
Growth Temperature 14 K

Pressurized at 30 atm

Journal of Low Temp. Phys. **111**, 497 (1998)

Crystal Growth from Gas Phase



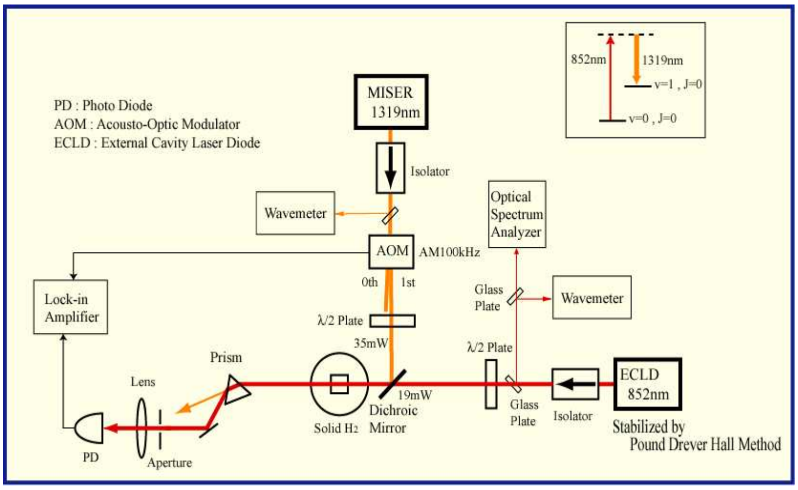


Growth Temperature 4~5 K

Substrate: Sapphire

Thickness < 500 μm

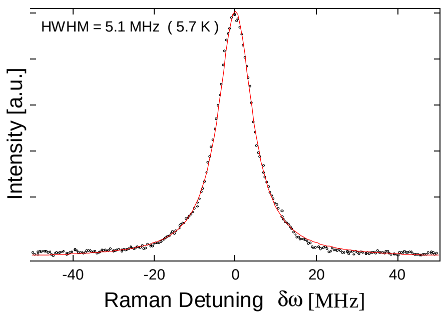
Raman Loss (Inverse Raman) Spectrometer



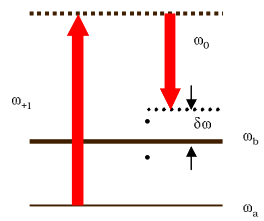
Spectral Resolution < 1 MHz (2 neV)

Coherent Raman Loss Spectrum

- Liquid Phase Growth -



$Q_1(0)$ Raman Transition



Crystal Thickness = 1 cm

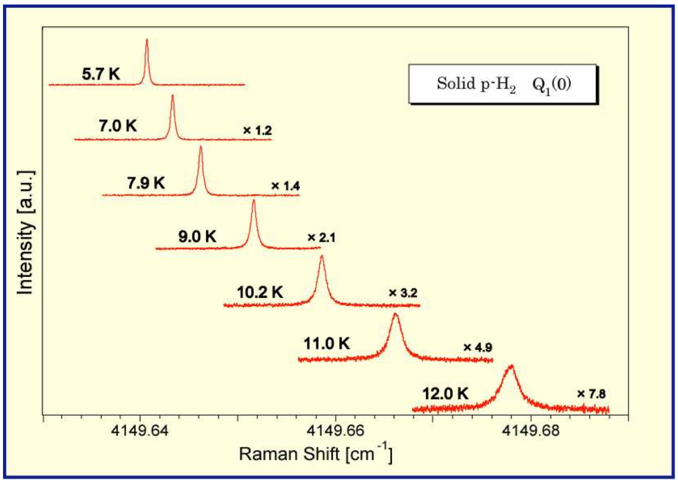
HWHM = 5.1 MHz (20 neV)

Ortho-H_2 Density ~ 0.01 %

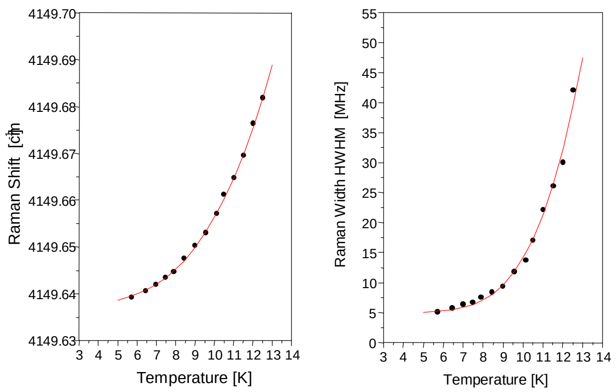
Raman Shift ~4150 cm^{-1}

Resolution < 1 MHz

Raman Spectrum of Solid Parahydrogen: Temperature Dependence



Temperature Dependence: Raman Shift & Width

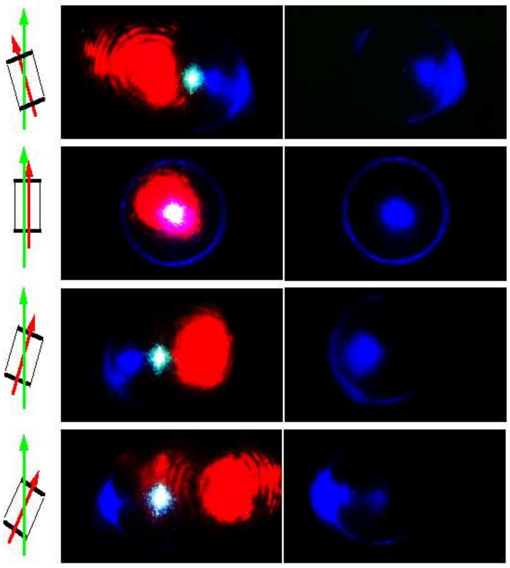


Black Dots: Experimental Observations

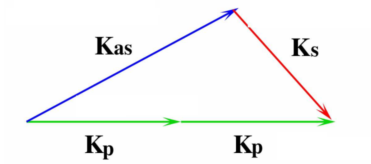
Red Curves: Theoretical Fitting through *Vibron-Phonon* Interaction

Stimulated Raman Scattering in Solid Hydrogen

- Self-Induced Phasmatching -



Traditional Phasematching



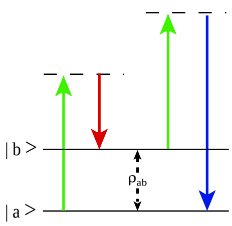
Green: **Pump Beam**

Red: **Stokes Beam**

Blue: **Anti-Stokes Beam**

Phys. Rev. Lett. **79**, 209 (1997)

Phil. Trans. R. Soc. Lond A **355**, 2405 (1997)



ρ_{ab}: . negligible dephasing

. Large Coherence . . . Large Nonlinearity

Maximum Conversion Length . . . . Phase Slip Length

"Maximum Conversion within One Coherence Length"

Parametric Process without Conventional Phasematching Restrictions

Highly Controlled "Large Coherence" .

Arbitrary & Efficient Frequency Modulation

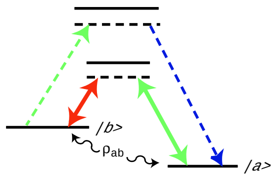
Quantum Crystal Oscillator/Modulator

Nonlinear Optics with Large Coherence

- EIT Nonlinear Optics: Frequency Converter -

Large ρ_{ab} in Dark State
  # Narrow Raman width
  # Gas phase low density system
  # Reasonably large Raman frequency

Resonant Raman Scheme

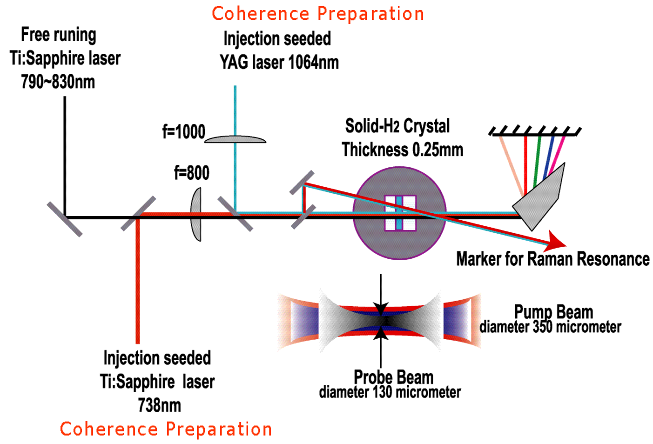


Extension to High Density Solid Medium

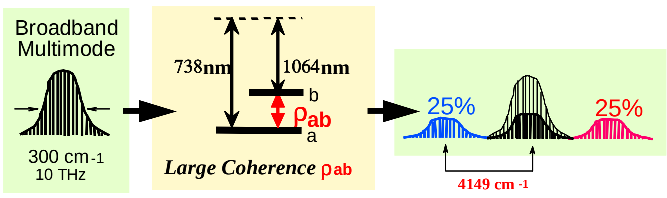
New Freedoms:
  # Far-off resonance system   "Broad bandwidth"
  # Short interaction length
  # Can manipulate boundary conditions

Key Point:
  Must keep the *"Narrow Raman Width"*

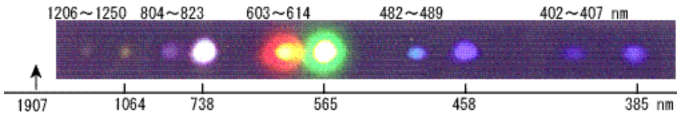
Parametric Raman Beating with a Broadband Laser



Arbitrary & Efficient Sideband Generation

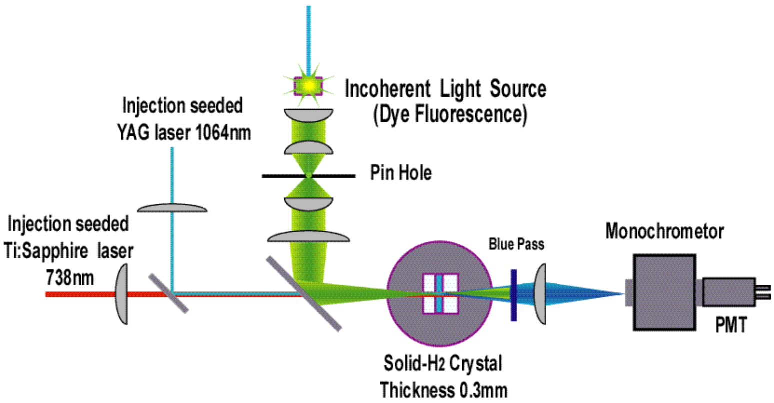


Parametric Raman Sideband Spectrum

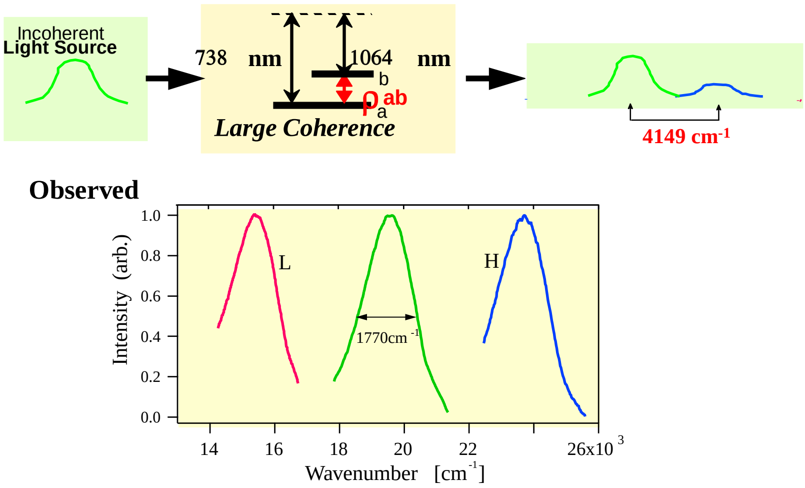


Phys. Rev. Lett. **85**, 2474 (2000)

Sideband Copying of Incoherent Light

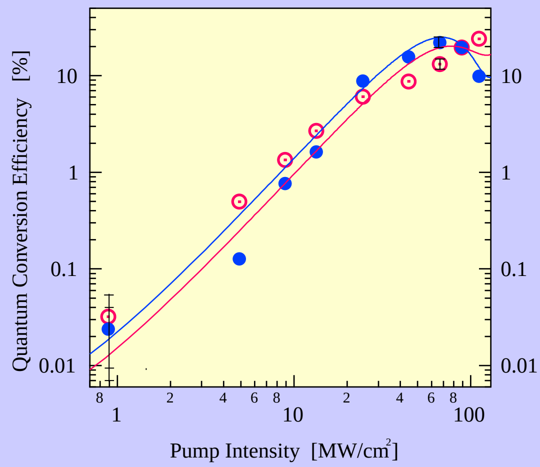


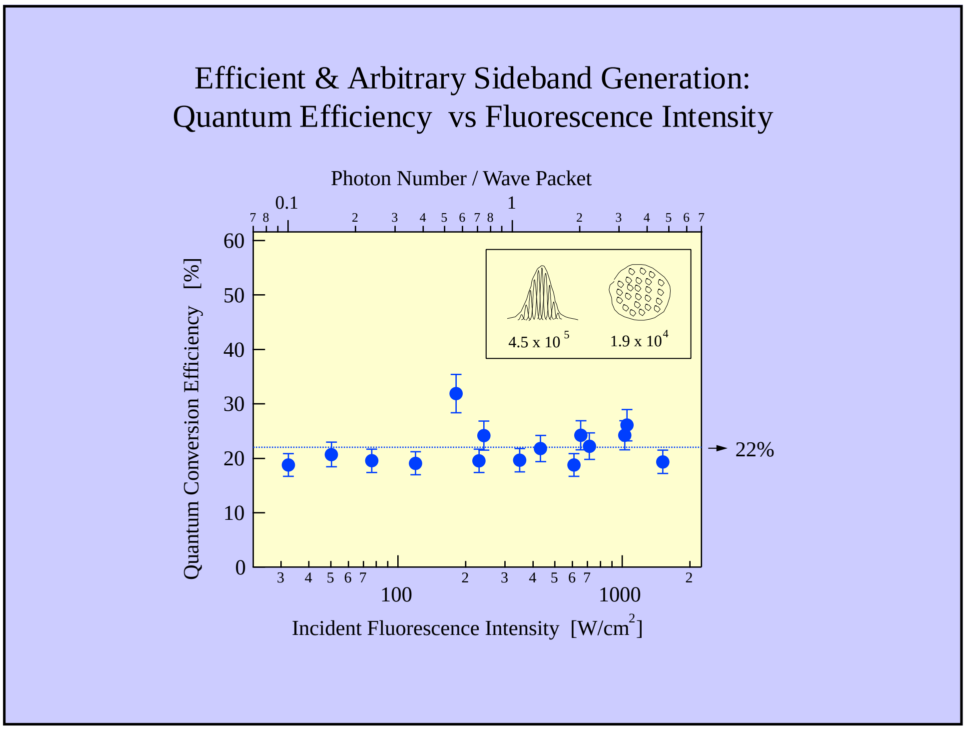
Sideband Copying of Incoherent Light :
- Spectrum Copying -



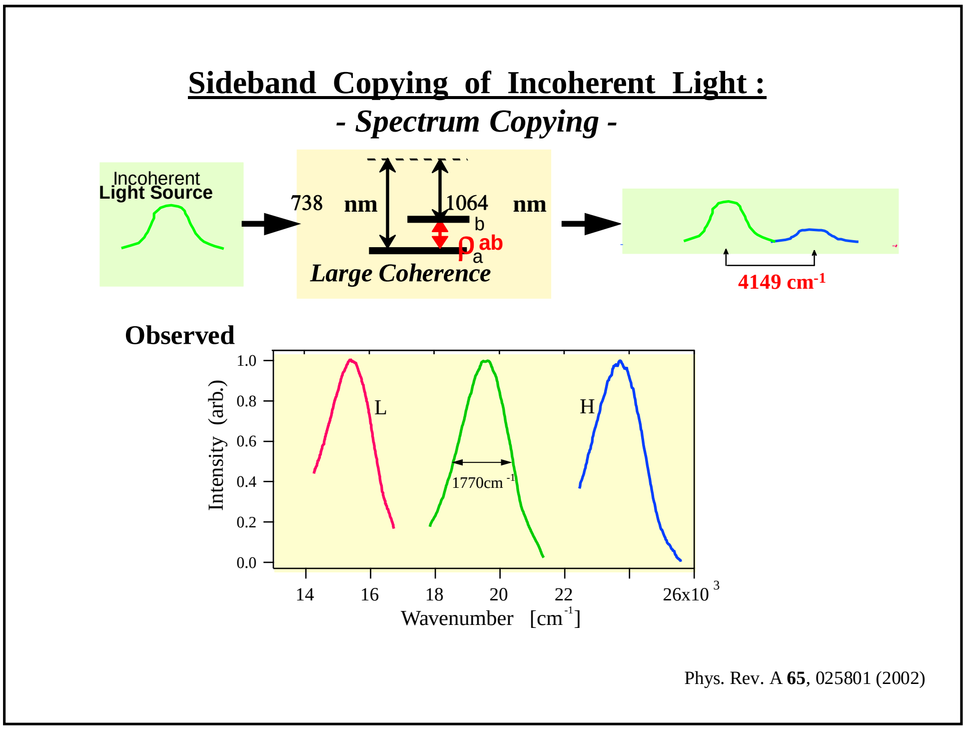
Phys. Rev. A **65**, 025801 (2002)

Efficient & Arbitrary Sideband Generation:
Quantum Efficiency vs Coherence Preparation





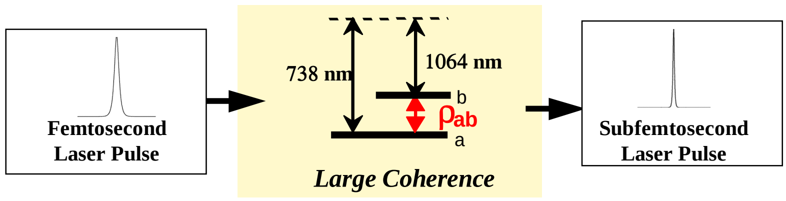
Efficient & Arbitrary Sideband Generation:
Quantum Efficiency vs Fluorescence Intensity
Photon Number / Wave Packet
0.1
1
4.5 x 10^5
1.9 x 10^4
22%
Quantum Conversion Efficiency [%]
100
1000
Incident Fluorescence Intensity [W/cm²]



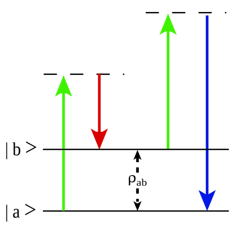
Sideband Copying of Incoherent Light :
- Spectrum Copying -
Incoherent Light Source
738 nm
1064 nm
Large Coherence
4149 cm⁻¹
Observed
L
H
1770cm⁻¹
Intensity (arb.)
Wavenumber [cm⁻¹]
Phys. Rev. A 65, 025801 (2002)

Possible Extension

- Generation of Subfemtosecond Light Pulse -



Phys. Rev. A **60**, 1562 (1999)



"Quantum Crystal Oscillator/Modulator"

Large Coherence with Strong Coupling

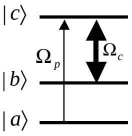
Highly Controlled "Large Coherence"
Arbitrary & Efficient Frequency Converter

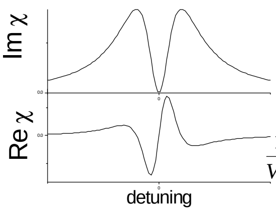
Small Coherence with Strong Coupling: *EIT*

Slow Light

Slow light, Frozen Light with Alkali Atoms

Electromagnetically Induced Transparency(EIT):
Strong Coupling in a Resonant Λ-System



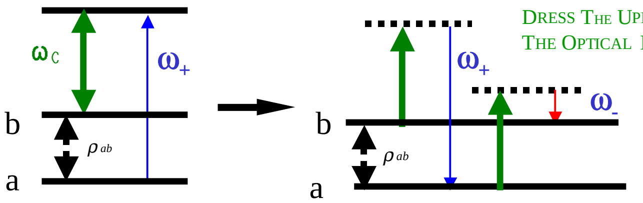


$$\frac{1}{V_G} - \frac{1}{c} = \frac{\omega}{2c}\frac{\partial\chi}{\partial\omega}$$

Can we realize the Slow Light in a transparent far-off resonance system?

STRONG COUPLING: SMALL COHERENCE

ONE STRONG FIELD :
DRESS THE UPPER STATES & CONTROL
THE OPTICAL RESPONSE FOR THE PROBE FIELD



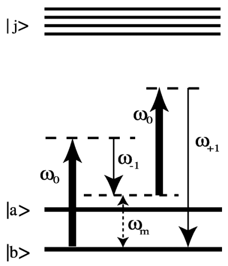
ρ_{ab} : SMALL

$\rho_{ab} \otimes \Omega_c @ \omega_c \rightarrow$ $\omega_+ : (\omega_c + \omega_{ab})$, $\omega_- : (\omega_c - \omega_{ab})$

COUPLING: STRONG STATIONARY FIELD
PROBE: WEAK PULSE FIELD
KEY: REALIZATION OF STEEP TWO-PHOTON DISPERSION
SLOW GROUP VELOCITY → LONG INTERACTION TIME
LARGE NONLINEARITY

Canad. J. Phys. **78**, 543 (2000)

Model & Basic Equations



$$H_{eff} = -\frac{\hbar}{2}\begin{bmatrix} \sum_q a_q |E_q|^2 & \sum_q d_q E_q E^*_{q+1} \\ \sum_q d^*_q E^*_q E_{q+1} & \sum_q b_q |E_q|^2 - 2\delta \end{bmatrix}$$

$$\frac{\partial E_q}{\partial z} = i\frac{N\hbar\omega_q}{\varepsilon_0 c}\left[\left(a_q\rho_{aa} + b_q\rho_{bb}\right)E_q + d_{q-1}\rho_{ba}E_{q-1} + d^*_q\rho_{ab}E_{q+1}\right]$$

Copropagating Two Sideband Fields

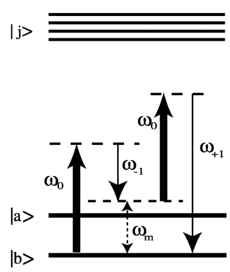
$$\left(\frac{\partial}{\partial z} - \frac{1}{V_{-1}}\frac{\partial}{\partial \tau}\right)E^*_{-1} = -i\left(\kappa_{-1}E^*_{-1} + \sigma_{-1}E_1\right) + \frac{1}{v_{-1}}\frac{\partial E_1}{\partial \tau}$$

$$\left(\frac{\partial}{\partial z} + \frac{1}{V_1}\frac{\partial}{\partial \tau}\right)E_1 = i\left(\kappa_1 E_1 + \sigma_1 E^*_{-1}\right) - \frac{1}{v_1}\frac{\partial E^*_{-1}}{\partial \tau}$$

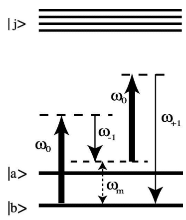
$$\frac{1}{V_1} = \frac{N\hbar\omega_1 d_0{}^2 E_0{}^2}{2\varepsilon_0 c\delta_e{}^2}$$

$$\frac{1}{V_{-1}} = \frac{N\hbar\omega_{-1} d_{-1}{}^2 E_0{}^2}{2\varepsilon_0 c\delta_e{}^2}$$

$$\frac{1}{v_{\pm 1}} = \frac{N\hbar\omega_{\pm 1} d_0 d_{-1} E_0{}^2}{2\varepsilon_0 c\delta_e{}^2}$$



Model & Basic Equations

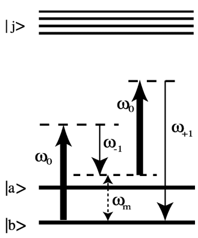


$$\frac{\partial E_q}{\partial z} = i\frac{N\hbar\omega_q}{\varepsilon_0 c}\left[\left(a_q\rho_{aa} + b_q\rho_{bb}\right)E_q + d_{q-1}\rho_{ba}E_{q-1} + d_q^*\rho_{ab}E_{q+1}\right]$$

Approximation

Negligible Dispersion: $a_q = a_0,\quad b_q = b_0,\quad d_q = d_0$

Slow Light in a Far-Off-Resonance System



$$\left(\frac{\partial}{\partial z} + \frac{\partial}{c\partial t}\right)E_{+1} = i\frac{N\hbar\omega_{+1}}{\varepsilon_0 c}d_0 E_0 \rho_{ba}$$

$$\left(\frac{\partial}{\partial z} + \frac{\partial}{c\partial t}\right)E_{-1}^* = -i\frac{N\hbar\omega_{-1}}{\varepsilon_0 c}d_0 E_0 \rho_{ba}$$

Propagation Normal Mode

$$E_u = \frac{\omega_{-1}E_{+1} + \omega_{+1}E_{-1}^*}{\omega_0}$$

$$\left[\frac{\partial}{\partial z} + \frac{\partial}{c\partial t}\right]E_u = 0$$

Propagate with Vacuum-Speed of Light

Dark-State Mode

$$E_v = E_{+1} + E_{-1}^*$$

$$\left[\frac{\partial}{\partial z} + \left(\frac{1}{c} + \frac{1}{V}\right)\frac{\partial}{\partial t}\right]E_v = i\frac{\delta_e}{V}E_v$$

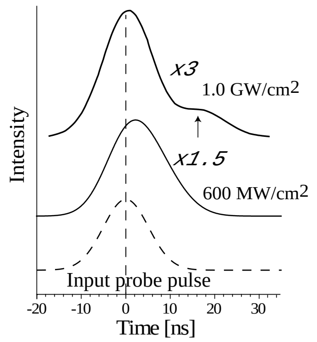
$$\frac{1}{V} = \frac{N\hbar\omega_m d^2 E_0^2}{\varepsilon_0 c \delta_e^2}$$

Propagate with Reduced Velocity

Bright-State Mode

Normal Mode Propagation for Slow Light

- Calculation for a Stokes Pulse -



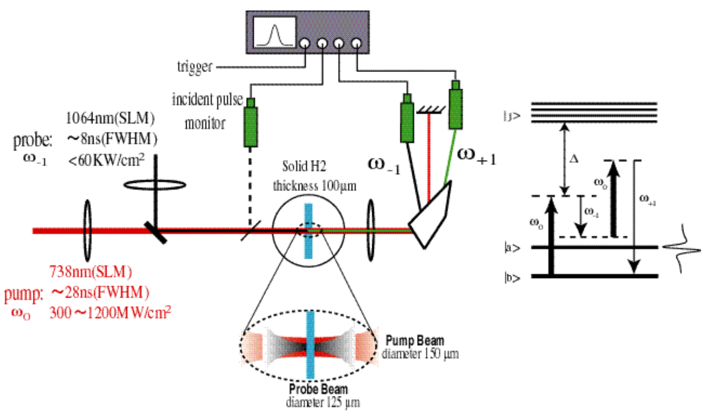
Raman Detuning = +70 MHz

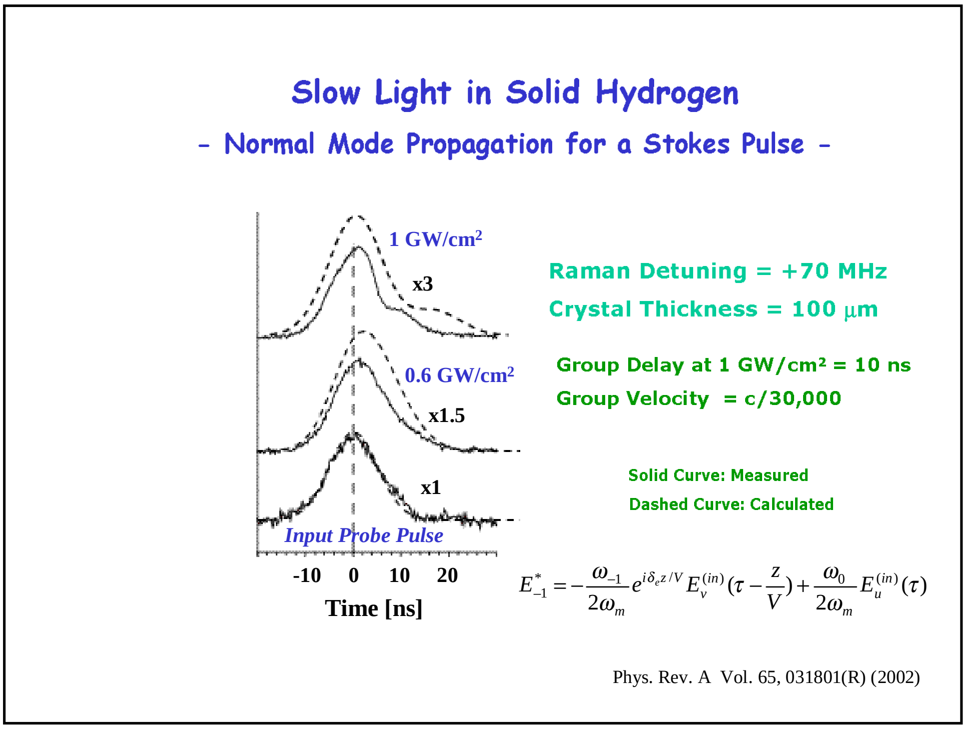
Crystal Thickness = 100 μm

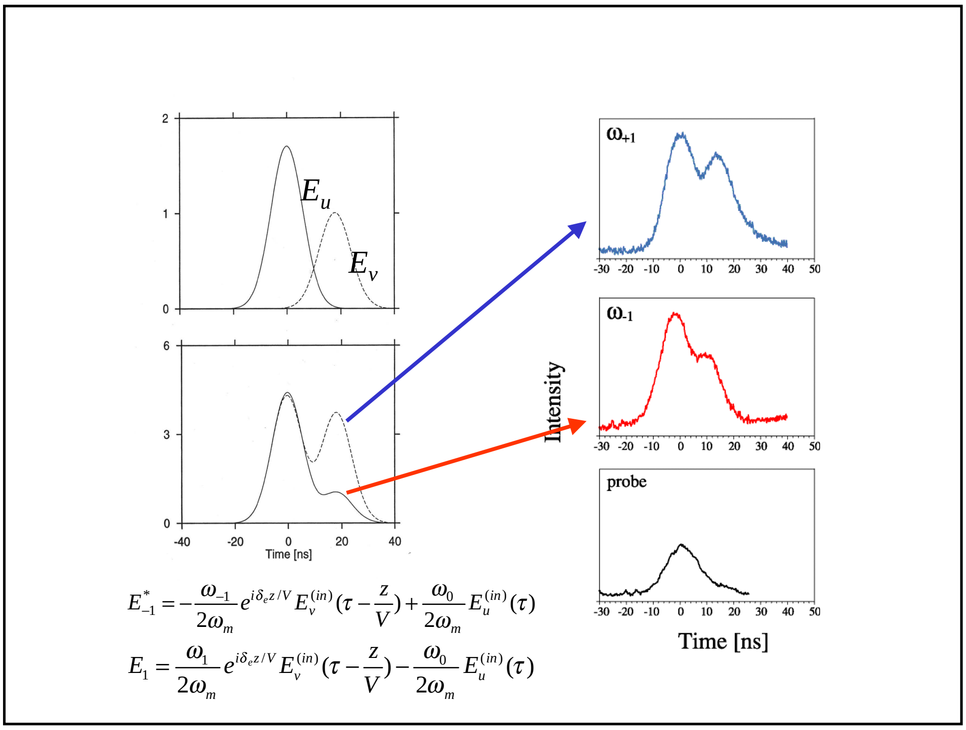
Stokes Field

$$E_{-1}^{*} = -\frac{\omega_{-1}}{2\omega_m} e^{i\delta_e z/V} E_v^{(in)}(\tau - \frac{z}{V}) + \frac{\omega_0}{2\omega_m} E_u^{(in)}(\tau)$$

Experimental setup




Slow Light in Solid Hydrogen
- Normal Mode Propagation for a Stokes Pulse -
1 GW/cm²
x3
0.6 GW/cm²
x1.5
x1
Input Probe Pulse
-10 0 10 20
Time [ns]
Raman Detuning = +70 MHz
Crystal Thickness = 100 μm
Group Delay at 1 GW/cm² = 10 ns
Group Velocity = c/30,000
Solid Curve: Measured
Dashed Curve: Calculated
$E_{-1}^{*} = -\frac{\omega_{-1}}{2\omega_m} e^{i\delta_e z/V} E_v^{(in)}(\tau - \frac{z}{V}) + \frac{\omega_0}{2\omega_m} E_u^{(in)}(\tau)$
Phys. Rev. A Vol. 65, 031801(R) (2002)


E_u
E_v
Time [ns]
ω_{+1}
ω_{-1}
probe
Intensity
Time [ns]
$E_{-1}^{*} = -\frac{\omega_{-1}}{2\omega_m} e^{i\delta_e z/V} E_v^{(in)}(\tau - \frac{z}{V}) + \frac{\omega_0}{2\omega_m} E_u^{(in)}(\tau)$
$E_1 = \frac{\omega_1}{2\omega_m} e^{i\delta_e z/V} E_v^{(in)}(\tau - \frac{z}{V}) - \frac{\omega_0}{2\omega_m} E_u^{(in)}(\tau)$

Slow Light in a Transparent Medium

Vibron Raman System in Solid Hydrogen
Propagation Normal Mode: Dark State Mode & Bright State Mode
Experimental Demonstration

Can the solid hydrogen work as a "resonant slow-light medium"?

Resonant EIT system in Solid Hydrogen

Doping Atoms, Molecules in Solid Hydrogen

Solid hydrogen as a Matrix:

Extremely Small Inhomogeneity

Orthohydrogen **as a Molecular Dopant**

Orthohydrogen: ***Nuclear Spin I = 1***

Vibrational Transition: ***Allowed via Crystal Field***

Transition Wavelength: 2.4 μm

EIT using nuclear-spin sublevels through vibrational transition

Nuclear Spin Manipulation via Molecular Vibration

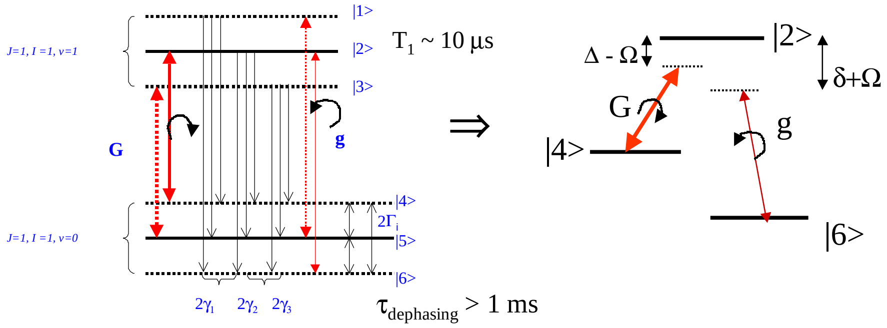
Slow Light, Frozen Light, Parametric Beating, *etc*

Resonant EIT system in Solid Hydrogen

Orthohydrogen as a Molecular Dopant

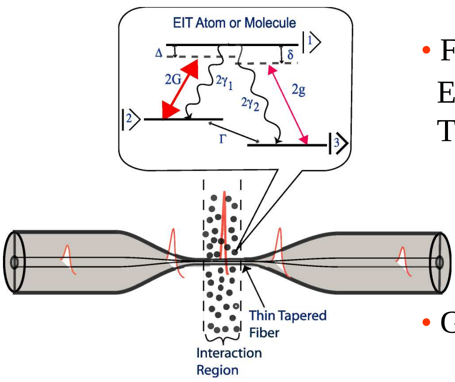
EIT using nuclear spin sublevels in a **B**-field through vibrational transition

Nuclear Spin Manipulation via Molecular Vibration



Transition Wavelength: around ~2.4 μm

Propagation through a Tapered Optical fiber



- Fiber mode can be *tailored* via Electromagnetically Induced Transparency (EIT)
- Group velocity of the fiber mode $v_g = [d\beta/d\omega]^{-1}$ ~ 44 m/s.

Evanescent-like field interacts with EIT Atoms / Molecules

Summary

Why Solid Hydrogen?

It is a Solid that meets both high-density and single-molecular nature.

How good is it?

- *High resolution Raman spectroscopy*

It shows an extremely narrow width of 5 MHz..

Nonlinear Optics in Solid Hydrogen

It works as an efficient and arbitrary "Quantum Crystal Modulator".

Slow Light

- *It slows down the pulse speed of light even in a transparent condition.*

Is it extendable?

It may also work as a resonant EIT medium.

Funding Support

CREST, Japan Science and Technology Corporation (JST)