Using Quantum Coherence for Remote Sensing

Marlan O. Scully

With A. Sokolov, P. Sprangle, and A. Svidzinsky



Texas A&M University

Princeton University





Paint Branch Distinguished Lecture in Applied Physics University of Maryland, October 2015

RAMAN AT RANGE

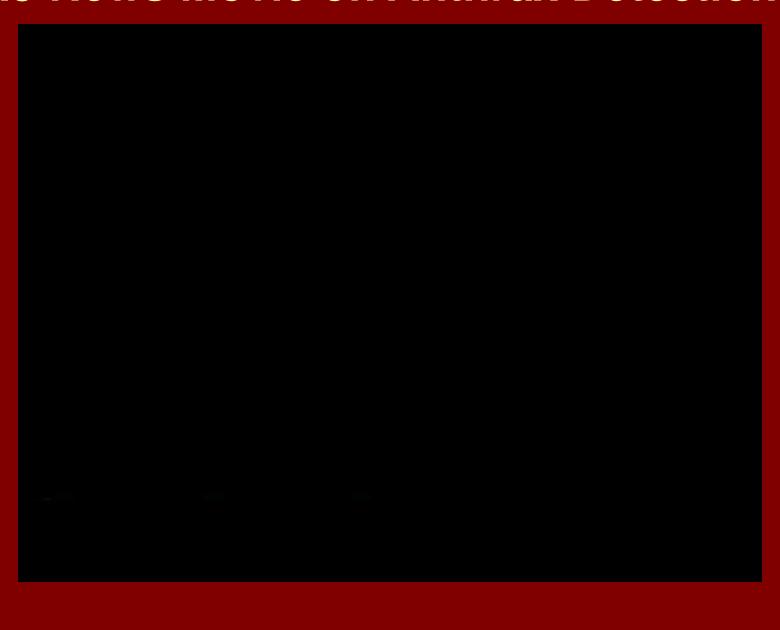


- 1) Anthrax Detection via FAST CARS
 - a) Motivation
 - b) Coherent vs. Spontaneous Raman (Boyd, Shen, Welch)
- 2) Random Raman
 - a) Remote Chemical Detection
 - b) Bone Density Measurements

DICKE AT A DISTANCE

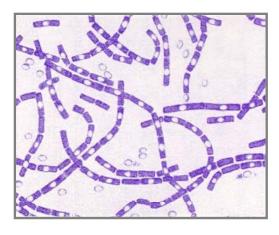
- 3) Superradiant Swept Gain
 - a) High Gain Backward Lasing
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 - a) Concept and Numerical Simulation
 - b) Simple Gain Calculation
- 5) CARS in the Sky
- 6) Summary

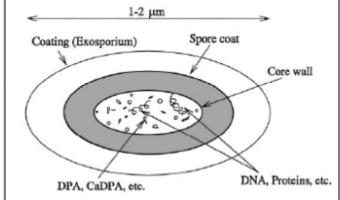
NJ News Movie on Anthrax Detection

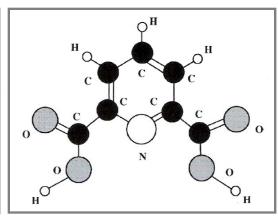


M.O. Scully et al., PNAS 99, 10994 (2002)

FAST CARS: Engineering a laser spectroscopic technique for rapid identification of bacterial spores







Bacillus anthracis http://textbookofbacteriology.net

Sketch of a spore M. O. Scully et al, PNAS, 2002

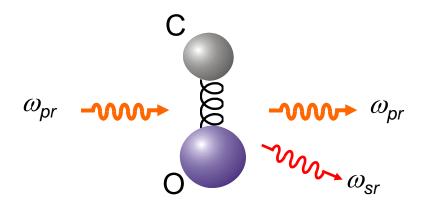
Dipicolinic acid Marker-molecule!

FAST CARS – Femtosecond Adaptive Spectroscopic Technique for Coherent Anti-Stokes Raman Scattering

- G. Beadie et al. (Washington, USA)
- M. Mehendale et al. (Princeton, USA)
- T. Siebert, W. Kiefer et al. (Wuerzburg, Germany)

Spontaneous Raman Scattering

Raman effect: experimentally discovered in February of 1928, by

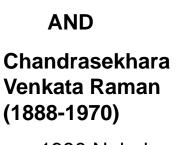


all predicted theoretically by Adolf Smekal in 1923! Leonid Isaakovich Mandelshtam (1879-1944)

Grigory Samuilovich Landsberg (1890-1957)



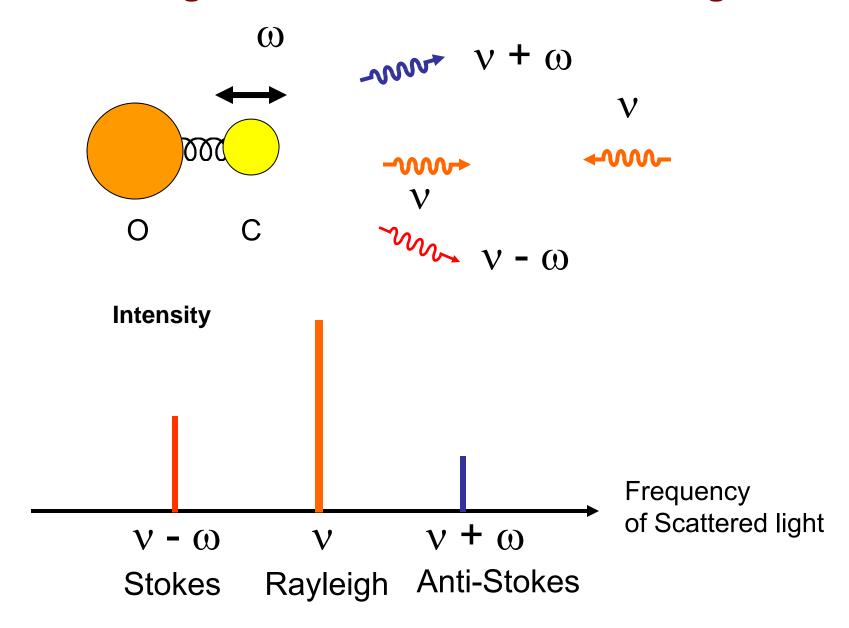




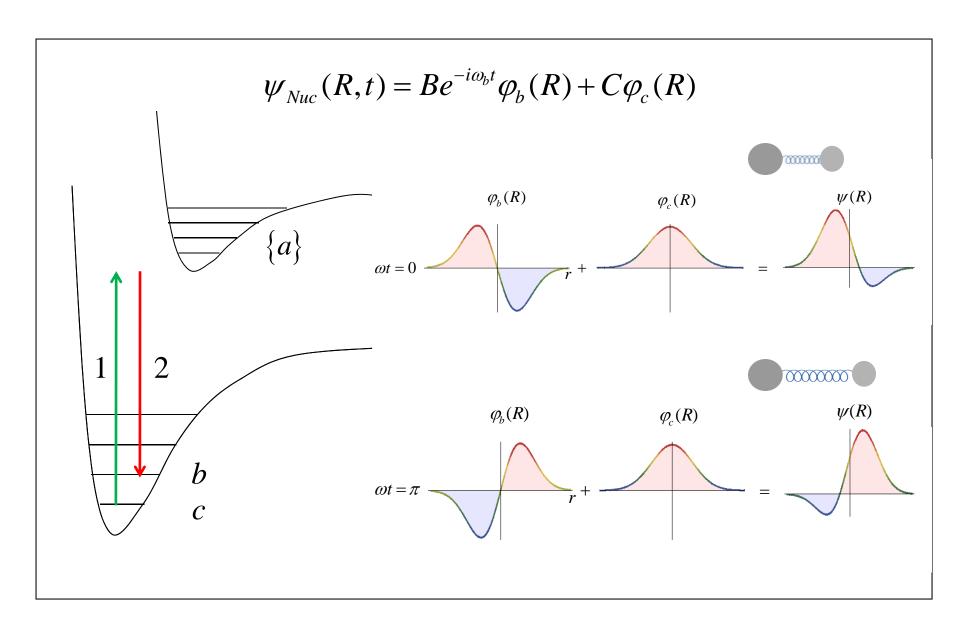
1930 Nobel in Physics



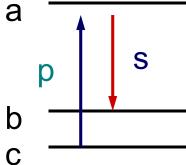
Raman Scattering is like Reflection off Oscillating Mirror



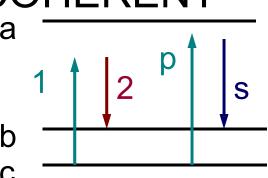
Raman Coherence, ρ_{bc} , implies Oscillation of Nuclear separation



INCOHERENT



COHERENT



Raman Hamiltonian

$$H_{p,s} = \sum_{i} G(|b\rangle\langle c|)_{j} a_{s}^{+} a_{p} e^{i(\vec{k}_{p} - \vec{k}_{s}) \cdot r_{j}} + adj.$$

Equation of motion for Stokes field

$$\dot{a}_{s} = -\frac{i}{\hbar} \left[H_{p,s}, a_{s} \right]$$

Short Pulse

$$a_s(\tau) \cong -\frac{i}{\hbar} \sum_j G \tau (|b\rangle\langle c|)_j a_s^+ a_p e^{i(\vec{k}_p - \vec{k}_s) \cdot r_j}$$

Number of Stokes photons

$$\langle n_s(\tau) \rangle \cong \sum_{i,j} \left(\frac{G\tau}{\hbar} \right)^2 \langle (|c\rangle\langle b|)_i (|b\rangle\langle c|)_j \rangle \langle n_p \rangle e^{i(\vec{k}_p - \vec{k}_s) \cdot (\vec{r}_j - \vec{r}_i)}$$

$$\langle n_{s}(\tau)\rangle \cong \left\{\sum_{i} \rho_{cc}^{i} + \sum_{i\neq j} \rho_{bc}^{i} \rho_{cb}^{j} e^{i\left[\vec{k}_{1} + \vec{k}_{p} - \vec{k}_{2} - \vec{k}_{s}\right] \cdot \left(\vec{r}_{j} - \vec{r}_{i}\right)}\right\} \left(\frac{G\tau}{\hbar}\right)^{2} \langle n_{p}\rangle$$

Spontaneous

Cooperative

Incoherent

Coherent

One Atom

N Atom

$$\langle n_s(\tau) \rangle \cong N \left(\frac{G\tau}{\hbar}\right)^2 \rho_{cc} + \frac{N(N-1)}{V} \left(\frac{G\tau}{\hbar}\right)^2 \left|\rho_{bc}\right|^2 \lambda^2 R$$

Incoherent Raman

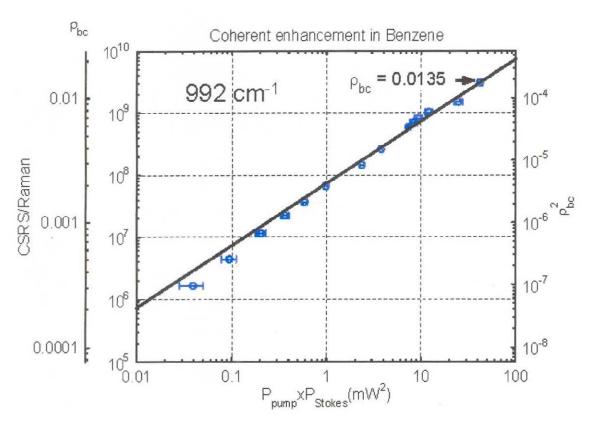
Coherent Raman

Bottom Line

$$\frac{\left\langle n_{s}\right\rangle_{coh}}{\left\langle n_{s}\right\rangle_{incoh}} \cong \frac{N}{V} \lambda^{2} R \frac{\left|\rho_{bc}\right|^{2}}{\rho_{cc}}$$

Cooperative Spontanous Emission

Raman "amplification" via coherence: Experimental coherence measurement



$$\frac{Coherent}{Incoherent} = \lambda^2 L \frac{N}{V} \frac{4\pi}{\Omega} \rho_{bc}^2$$

$$\rho_{bc}$$
 , experimental = 0.0135 ρ_{bc} , theoretical = 0.018

- 9-10 orders of magnitude enhancement over spontaneous Raman
- High directionality
- Maximum theoretical coherence: 1/2



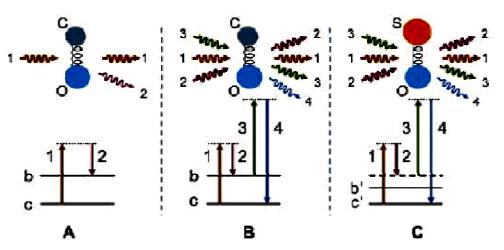
Science

SCIENCE VOL 316 13 APRIL 2007

Optimizing the Laser-Pulse Configuration for Coherent Raman Spectroscopy

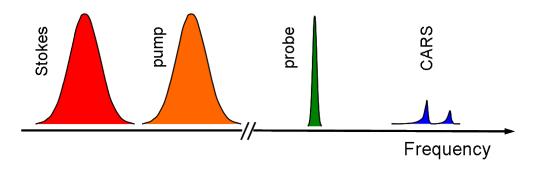
Dmitry Pestov, ¹* Robert K. Murawski, ^{1,2} Gombojav O. Ariunbold, ¹ Xi Wang, ¹ Miaochan Zhi, ¹ Alexei V. Sokolov, ¹ Vladimir A. Sautenkov, ¹ Yuri V. Rostovtsev, ^{1,2} Arthur Dogariu, ² Yu Huang, ² Marlan O. Scully ^{1,2}

Fig. 1. Level diagram and schematic of different scattering processes on simple molecules. In this example, CO is a target molecule and SO is a background molecule. (A) Incoherent Raman scattering (pulse 2) was derived from laser pulse 1 scattering off of the CO molecule. (B) CARS signal 4 was derived from probe

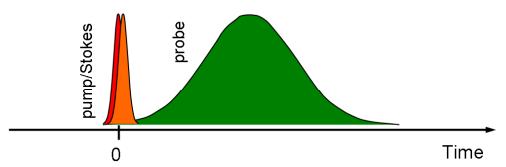


pulse 3 scattering off of the CO molecular vibration, coherently prepared by pulses 1 and 2. (C) One of the possible channels for the NR background generation in SO. c, the ground state of the CO molecule; b, the target vibrational state of the CO molecule; c', the ground state of the background molecule; b', an off-resonant vibrational state of the background molecule.

Optimal CARS Technique



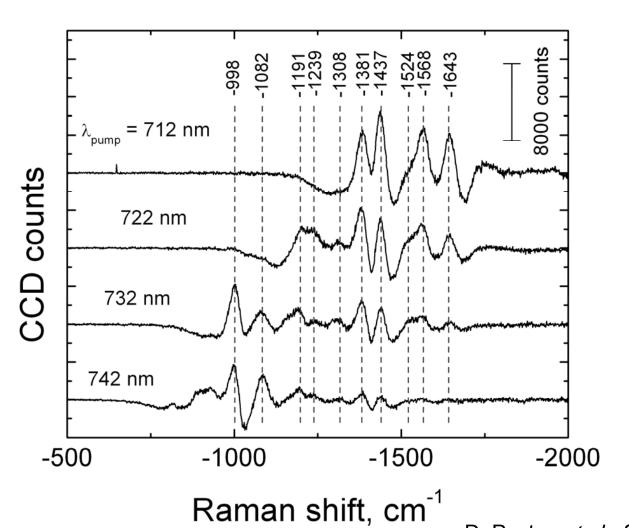
© Efficient broadband excitation and frequency-resolved probing, multi-channel detection => comprehensive species-specific information.



© The technique is relatively insensitive to signal amplitude fluctuations.

 Mitigation of non-resonant FWM and straightforward discrimination of the resonant response against the non-resonant one.

Backscattered CARS on spores



Parameters:

Pump 712-742 nm, 2.0 μJ/pulse

Stokes 803 nm, 3.9 µJ/pulse

Probe 577.9 nm, FWHM~0.7 nm ~0.5 µJ/pulse

Int. time is 2 min.

D. Pestov et al., Science **316**, 265 (2007).

Take-Home Message

FAST CARS is a promising technique that has a great potential for real-time detection and chemically-selective imaging applications.

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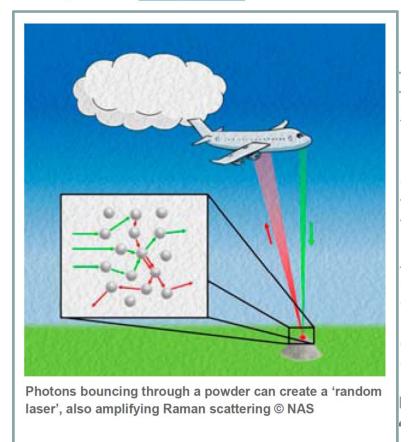
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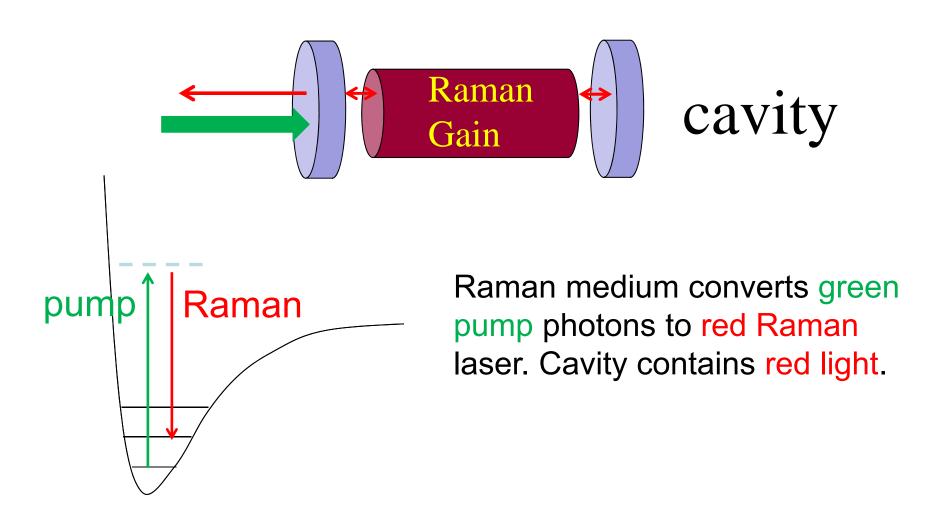
Photon pinball identifies chemicals from afar

13 August 2014 Andy Extance

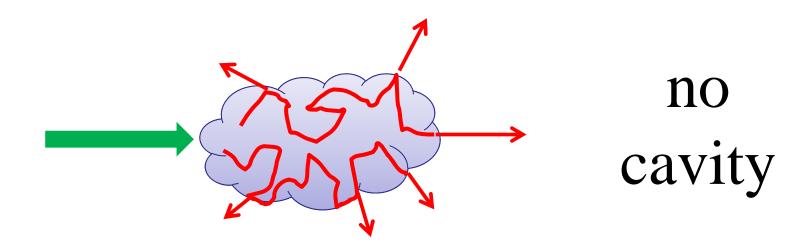


US scientists have pushed the range at which chemicals can be remotely identified beyond a kilometre by turning the samples themselves into lasers. By triggering bright random Raman laser emission, Marlan Scully and Vladislav Yakovlev from Texas A&M University and their team successfully distinguished a series of similar white powders.

What is a **Raman laser**?

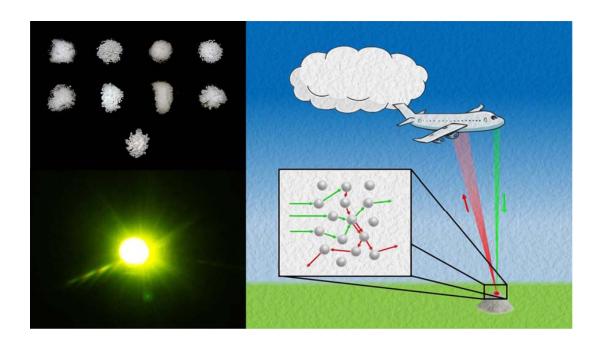


What is a random Raman laser?



Raman powder converts green pump photons to red Raman laser.
Scattering powder matrix contains red light.

Random Raman Laser



TAMU Team: 'Single-shot stand-off chemical identification of powders using random Raman lasing', News release.

The ability to remotely detect chemicals in real time at large distances opens the door to a variety of applications ranging from explosives monitoring and detection to monitoring nitrate levels for smart agriculture.

Spatially offset Raman microspectroscopy of highly scattering tissue: theory and experiment

Z. Di, B. Hokr, et al., (TBP)

Pioneering research by Matousek et al (Applied Spectroscopy, 2005) has demonstrated the feasibility of obtaining the density of human bones in vivo using Raman Random spectroscopy. The present work extends the measurements and uses Monte Carlo simulations to further demonstrate the effect.

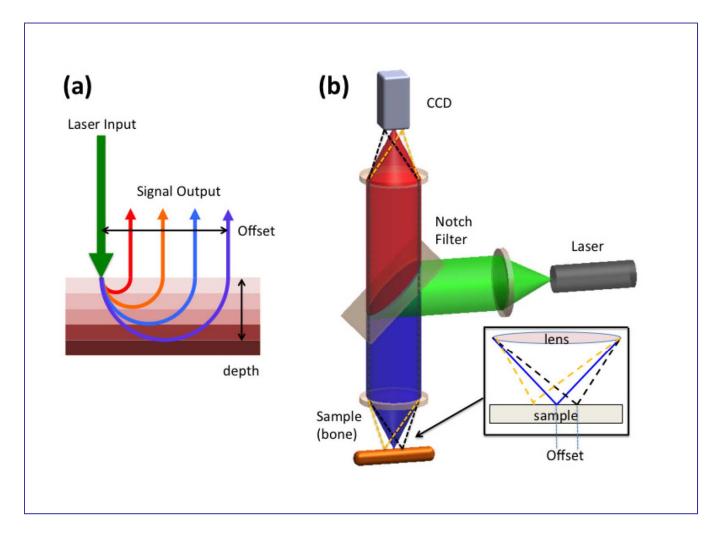


FIG. 1: Conceptual figure for (a): spatially offset Raman spectroscopy (SORS); (b): schematic diagram of the experimental microscope Raman detection setup.

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Standoff spectroscopy via remote generation of a backward-propagating laser beam

Philip R. Hemmer, et al., Texas A&M University

In an earlier publication we demonstrated that by using pairs of pulses of different colors (e.g., red and blue) it is possible to excite a dilute ensemble of molecules such that lasing and/or gain-swept superradiance is realized in a direction toward the observer.

In the present paper, we propose a related but simpler approach on the basis of the backward-directed lasing in optically excited dominant constituents of plain air, N_2 and O_2 .

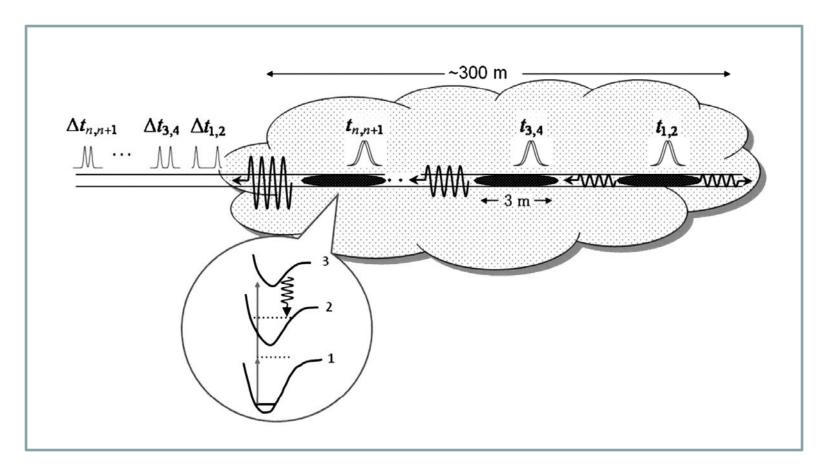


Fig. 1. SOS. Multiple pairs of pulses are generated such that the spacing between pulses in each pair is decreasing. The second pulse in each pair has a higher velocity because of atmospheric dispersion. The first pair of pulses overlaps near the back of the cloud, creating a small region of gain. Subsequent pairs overlap at closer and closer regions of the cloud, producing a swept-gain amplifier that lases back toward the observer.



High-Gain Backward Lasing in Air

Arthur Dogariu, et al., Princeton University

The compelling need for standoff detection of hazardous gases and vapor indicators of explosives has motivated the development of a remotely pumped, high-gain air laser that produces lasing in the backward direction and can sample the air as the beam returns. We demonstrate that high gain can be achieved in the near-infrared region by pumping with a focused ultraviolet laser.

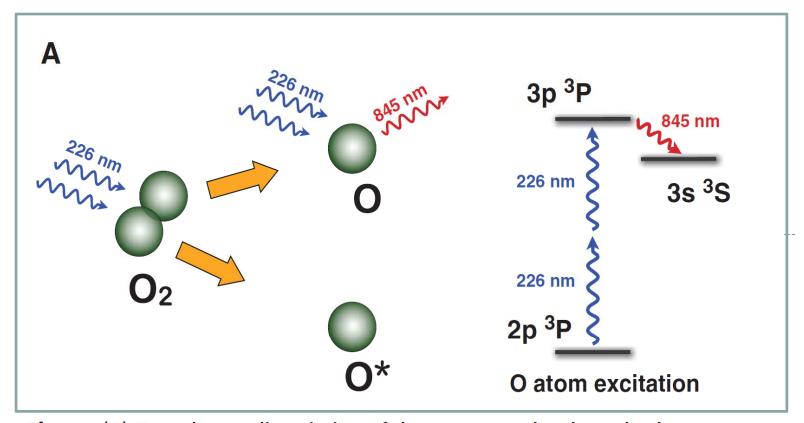


Fig. 1. (**A**) Two-photon dissociation of the oxygen molecule and subsequent two-photon resonant excitation of the ground-state oxygen atom fragment result in emission at 845 nm.



Coherence brightened laser source for atmospheric remote sensing*

Andrew Traverso, et al., Texas A&M University

"Our results suggest that the emission process exhibits atomic coherence (Dicke superradiance) in contrast with ordinary lasing where atomic coherence is negligible."

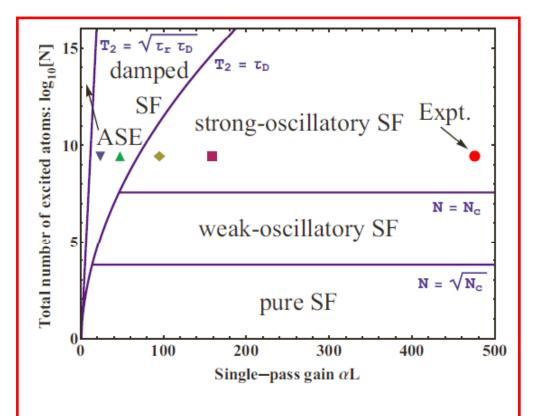


FIG. 1. (Color online) Regimes of *N*-atom cooperative spontaneous emission (adapted from Boyd et al.).

^{*}PNAS, **109**(38),15185-15190 (2012)

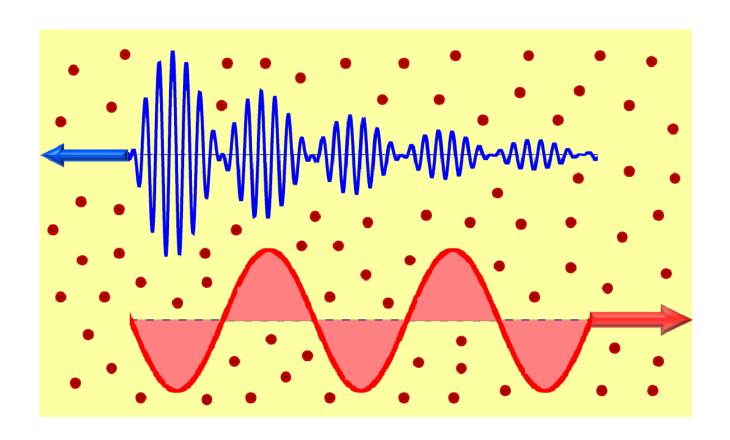
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Quantum Amplification by Superradiant Emission of Radiation (QASER)



A.A. Svidzinsky, L. Yuan, M.O. Scully, PHYSICAL REVIEW X 3, 041001, 2013

Quantum Amplification by Superradiant Emission of Radiation

Anatoly A. Svidzinsky, ^{1,2} Luqi Yuan, ^{1,2} and Marlan O. Scully ^{1,2,3}

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²Princeton University, Princeton, New Jersey 08544, USA

³Baylor University, Waco, Texas 76706, USA

(Received 18 June 2013; revised manuscript received 31 July 2013)

A laser generates light through stimulated emission of radiation and requires population inversion. Quantum interference can yield lasing without inversion. However, such phase-sensitive quantum amplification still requires some atomic population in the excited state. Here, we present a new kind of quantum amplifier based on superradiant emission of radiation (QASER) which does not need any population in the excited state!



SCIENCE

1510 18 SEPTEMBER 2009 VOL 325

PHYSICS

The Super of Superradiance

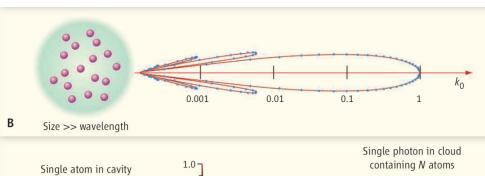
Marlan O. Scully^{1,2} and Anatoly A. Svidzinsky¹

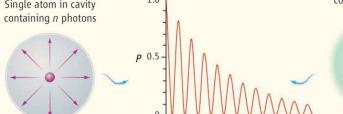
Cooperative single-photon emission from an atom ensemble will provide insights into quantum electrodynamics and applications in quantum communication.

Timed Dicke state

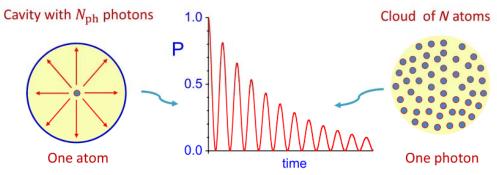
$$|\Psi\rangle = \frac{1}{\sqrt{N}} \left[e^{i\vec{k}_0 \cdot \vec{r}_1} \Big| \uparrow_1 \downarrow_2 \dots \downarrow_N \right\rangle + e^{i\vec{k}_0 \cdot \vec{r}_2} \Big| \downarrow_1 \uparrow_2 \dots \downarrow_N \right\rangle + \dots + e^{i\vec{k}_0 \cdot \vec{r}_N} \Big| \downarrow_1 \downarrow_2 \dots \uparrow_N \right\rangle$$







Size >>pulse length



	Probability of atomic excitation	Key variables V and N
Single atom maser	$sin^2 \frac{\wp}{\hbar} \sqrt{\frac{\hbar v}{\varepsilon_0 V}} \sqrt{N_{ph}} t$	$V =$ photonic cavity volume $N_{ph} =$ number of photons in cavity
Single photon superradiance	$sin^2 \frac{\wp}{\hbar} \sqrt{\frac{\hbar \nu}{\varepsilon_0 V}} \sqrt{N_{at}} t$	V= atomic cloud volume N_{at} = number of atoms in cloud

Can rewrite the collective superradiant oscillation frequency as:

$$\frac{\mathcal{O}}{\sqrt{\hbar\varepsilon_0}}\sqrt{\frac{\hbar\nu}{\varepsilon_0V}}\sqrt{N_{at}} = \sqrt{\frac{3}{4\pi}\lambda^2\gamma c\frac{N_{at}}{V}} \equiv \Omega_a$$

Defining the Rabi frequency $\Omega_s(z,t) = \wp \varepsilon_s(z,t)/\hbar$ we have our working equation:

$$\left(\frac{\partial}{\partial t} + c \frac{\partial}{\partial z}\right) \Omega_s = i \Omega_a^2 \rho_{ab}$$

Physics of QASER

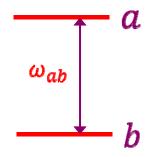
Electromagnetic field and atoms are two coupled oscillators

Maxwell's equation for electric field

$$\left(\frac{\partial^2}{\partial t^2} - c^2 \Delta\right) \Omega = -2 \frac{\Omega_a^2}{\omega_{ab}} \frac{\partial^2 \rho_{ab}}{\partial t^2}$$

Schrodinger equation for atoms

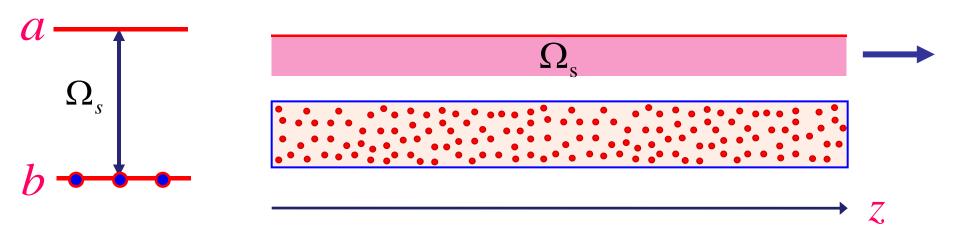
$$\frac{\partial \rho_{ab}}{\partial t} + i\omega_{ab}\rho_{ab} = i\Omega \left(\rho_{bb} - \rho_{aa}\right)$$



Coupling between oscillators is nonlinear

Driving of atoms modulates coupling strength

Light amplification with no population in excited state



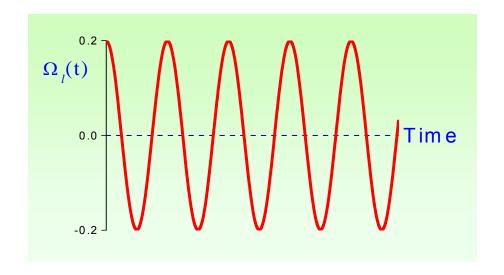
For uniform lasing field we obtain:

$$\frac{\partial^2 \Omega_s}{\partial t^2} + \Omega_a^2 (\rho_{bb} - \rho_{aa}) \Omega_s = 0$$

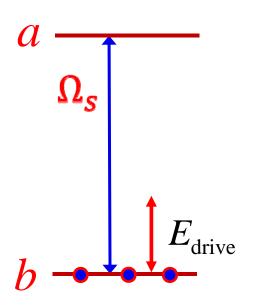
$$\Omega_a = \sqrt{\frac{3cN\lambda^2\gamma}{8\pi}}$$

Laser field oscillates with collective frequency:

$$\Omega_s(t) = \Omega_s(0)\cos\left(\Omega_a\sqrt{\rho_{bb}-\rho_{aa}}t\right)$$



What if we periodically change population of level a or b?



$$\frac{\partial^2 \Omega_s}{\partial t^2} + \Omega_a^2 (\rho_{bb} - \rho_{aa}) \Omega_s = 0$$

Mathieu equation

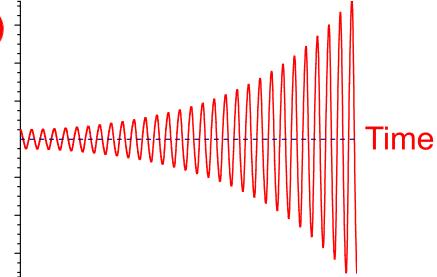
$$\frac{E_{\text{drive}}}{\partial t^2} + \Omega_a^2 \left[1 - \delta \cos(2\nu_d t) \right] \Omega_s = 0 \quad \delta << 1$$

If $v_d = \frac{\Omega_a}{m}$, m=1,2,3,... then field would exponentially grow with time:

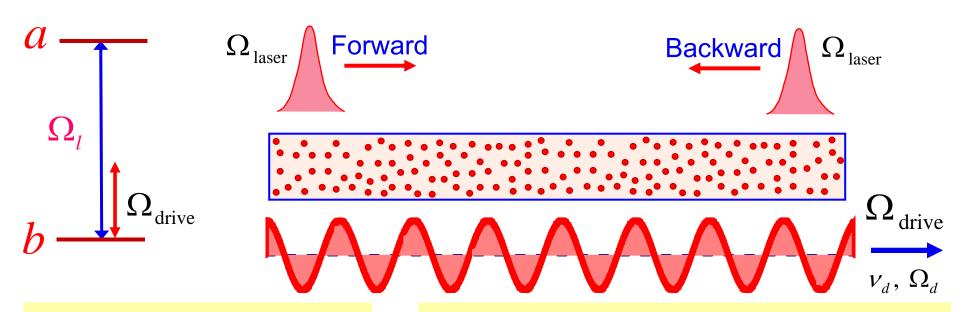
For 1st order resonance:

$$\Omega_s(t) \propto e^{Gt}, \quad G = \frac{\delta \cdot \Omega_a}{8}$$

G – gain per unit time



Propagating driving field: general analysis



Forward direction:



Backward direction:

If Stark shift is suppressed then

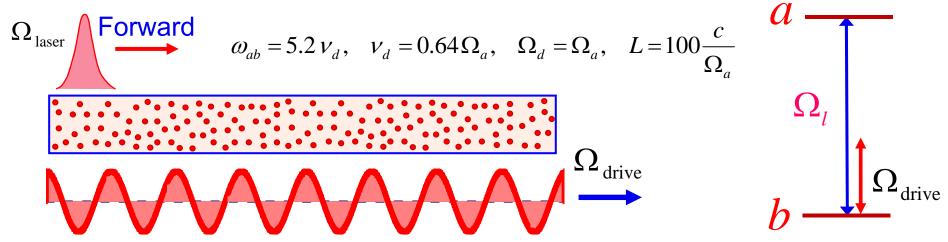
there is gain for $v_d > \frac{\Omega_a}{\sqrt{2}}$

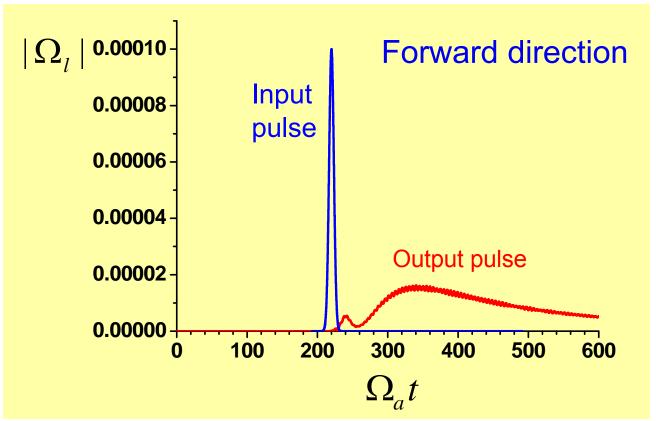
$$v_d > \frac{\Omega_a}{\sqrt{2}}$$

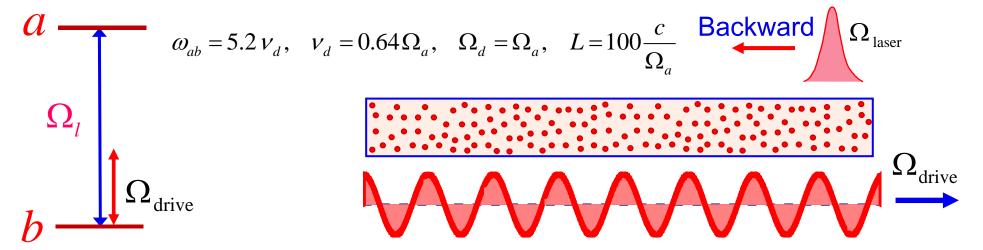
Gain per unit time:

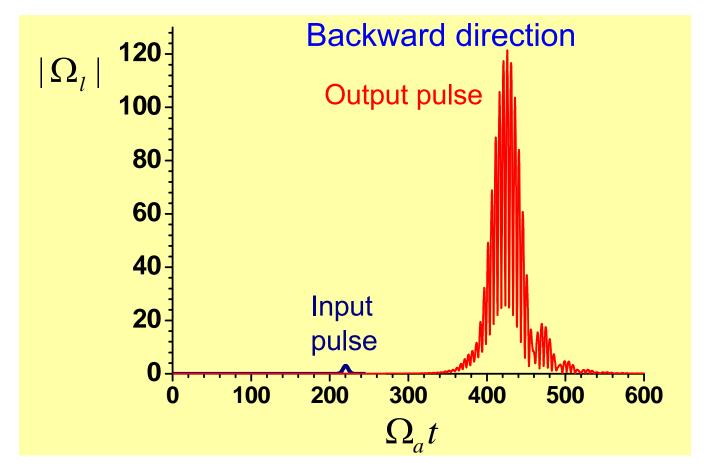
$$G_t = \frac{\Omega_a}{3\sqrt{2}} \left(\frac{\Omega_d}{\omega_{ab}}\right)^2$$

Numerical solution of Maxwell-Schrödinger equations



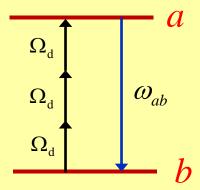






Comparison of atomic excitation mechanisms

Multiphoton resonant excitation



Single atom phenomenon

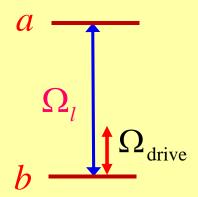
Multiphoton resonance with atomic transition frequency

$$\nu_d = \frac{\omega_{ab}}{m}$$

$$\rho_{aa} \approx \left(\frac{\Omega_d}{\omega_{ab}}\right)^{2m} (\omega_{ab} t)^2$$

High frequency light is emitted in the direction of driving field

Collective parametric resonance



Collective effect

Resonance with collective frequency

$$v_{d} > \frac{\Omega_{a}}{\sqrt{2}}$$

$$\rho_{aa} \approx \left| \rho_{ab}(0) \right|^{2} \exp \left(\frac{\sqrt{2}\Omega_{d}^{2}}{3\omega_{ab}^{2}} \Omega_{a} t \right)$$

Lasing occurs in the backward direction

THE QASER REVISITED:

Insights gleaned from analytical solutions to simple models*

Marlan O. Scully

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Texas A&M University,

College Station, TX 77843,

Baylor University, Waco, TX 76798

Lasers and masers typically require population inversion. But with phase coherent atoms(phasers), we get lasing without inversion (e.g., 10% of the atoms excited). However in recent work we found that it is possible to get coherent light emitted with no atoms excited, via Quantum Amplification of Superradiant Emission of Radiation(QASER). In particular, we found that utilizing collective superradiant emission, we can generate coherent light at high frequency in the UV or x-ray bands by driving the atomic system with a low-frequency (e.g., infrared) source. We here present a simple analysis based on near resonant QASER operation and on a multiphoton Hamiltonian obtained by, e.g., a canonical transformation.

SIMPLE GAIN EQUATION

$$\begin{array}{c|c}
\Delta & a \\
\Omega_{d} & \Omega_{s} \\
b
\end{array}$$

$$\dot{\Omega}_{s} = -i\Omega_{a}^{2}\rho_{ab}$$

$$\dot{\rho}_{ab} = -i\Omega_{s}(1-2\rho_{aa})$$

$$\dot{\rho}_{ab} = -i\Omega_{s}(1-2\rho_{aa})$$

$$\dot{\rho}_{ab} = (-i)^{2}\Omega_{a}^{2}(1-2\rho_{aa})\rho_{ab}$$

$$\ddot{X} = -\Omega_a^2 \left(1 - \left(\frac{\Omega_d}{\Delta} \right)^2 \cos 2\Delta t \right) X$$

$$X = \Omega_s \text{ or } \rho_{ab}$$

SAME EQUATION SAME GAIN

$$Gain = \frac{\Omega_a}{4} \left(\frac{\Omega_d}{\Delta} \right)^2$$

Parametric harmonic oscillator (Mathieu equation)

$$\frac{\partial^2 x}{\partial t^2} + \Omega_a^2 \left[1 - \delta \cos(v_d t) \right] x = 0 \qquad \delta << 1$$

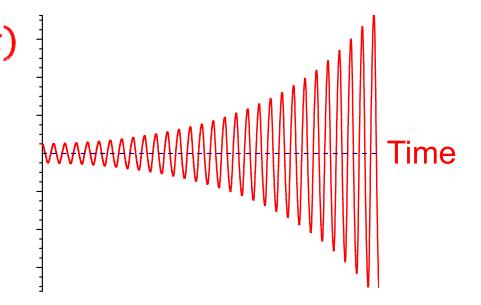
Parametric resonance

If
$$v_d = \frac{2\Omega_a}{m}$$
, $m=1,2,3,...$ then oscillations exponentially grow with time:

For 1st order resonance:

$$x(t) \propto e^{Gt}, \quad G = \frac{\delta \cdot v_d}{8}$$

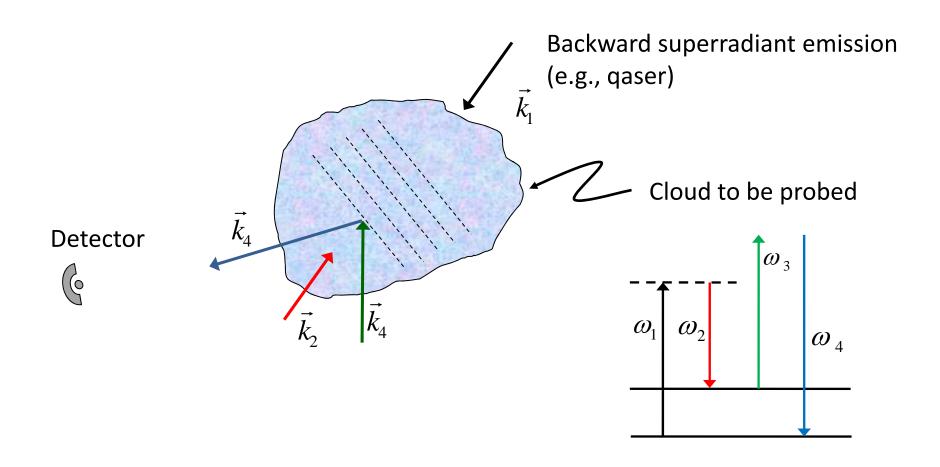
G – gain per unit time



QASER Summary

We found a new way to achieve light amplification in the backward direction with no population in excited state by means of collective parametric resonance.

CARS In The Sky*



^{*}Yuan, et al., Laser Phys. Lett. (2011)

"SUMMARY"

RAMAN AT RANGE

- 1) Anthrax Detection via FAST CARS
 - a) Motivation
 - b) Coherent vs. Spontaneous Raman (Boyd, Shen, Welch)
- 2) Random Raman
 - a) Remote Chemical Detection
 - b) Bone Density Measurements

DICKE AT A DISTANCE

- 3) Superradiant Swept Gain
 - a) High Gain Backward Lasing
 - b) Coherence Brightened Air Laser
- 4) Backward Quantum Amplification by Superradiant Emission of Radiation
 - a) Concept and Numerical Simulation
 - b) Simple Gain Calculation
- 5) CARS in the Sky (via S. L. Chin on the ground)
- 6) Summary

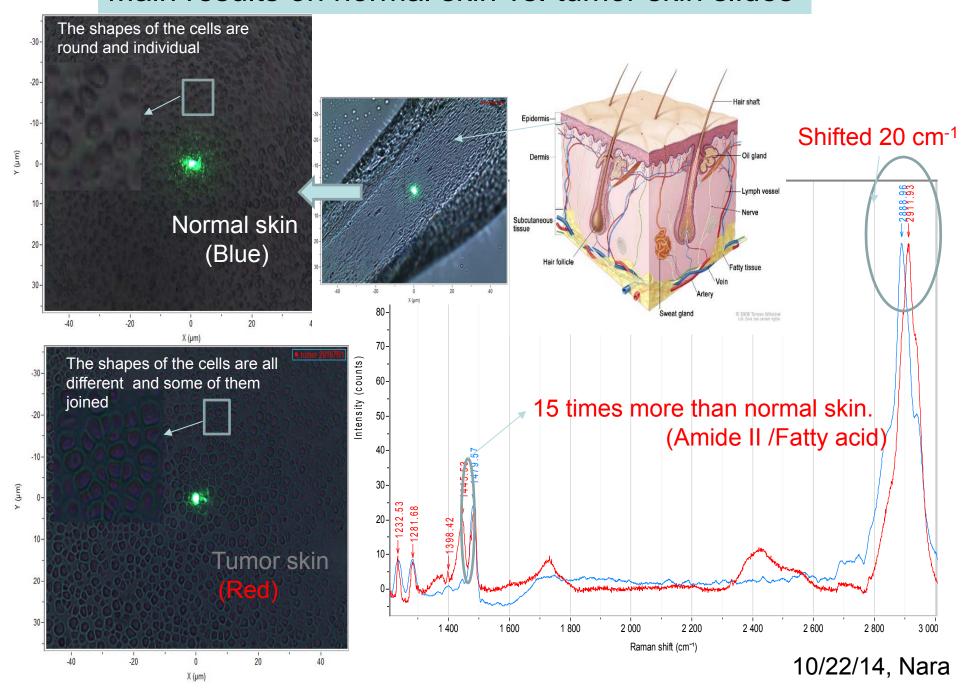
$$\frac{\left\langle n_{s}\right\rangle _{coh}}{\left\langle n_{s}\right\rangle _{incoh}}\cong\frac{N}{V}\lambda^{2}R\frac{\left|\rho_{bc}\right|^{2}}{\rho_{cc}}$$

 $E_{\rm out} \approx E_{\rm in} \exp \left(\frac{\Omega_d^2}{\Lambda^2} \Omega_a t \right)$

A Raman Spectroscopic Study of skin tumors of Sinclair Piglets



Main results on normal skin vs. tumor skin slides

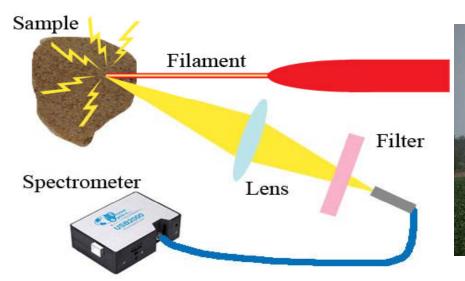


Filament-induced breakdown

spectroscopy (FIBS)

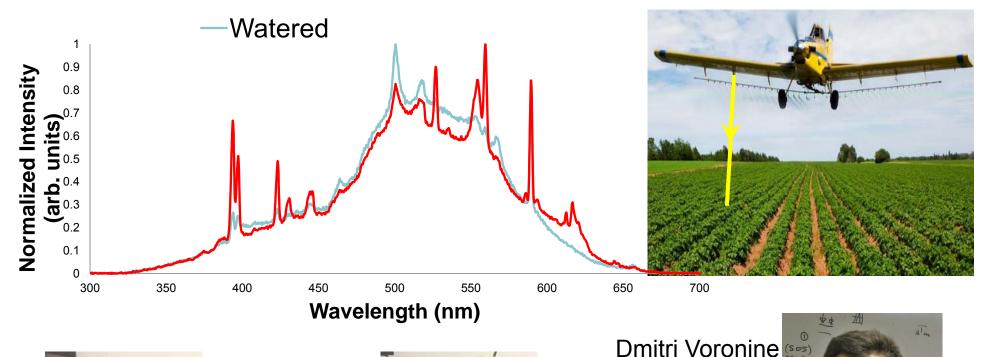
 Applications in precision agriculture, military, all areas of remote sensing

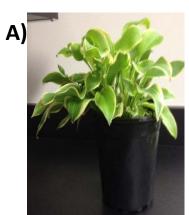
Remotely detect and identify biological and mineralogical samples



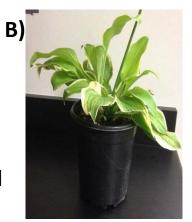


Laser-Induced Breakdown Spectroscopy (LIBS) of Hosta Leaf

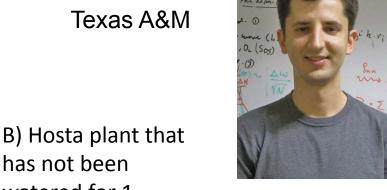




A) Hosta plant that is watered regularly.

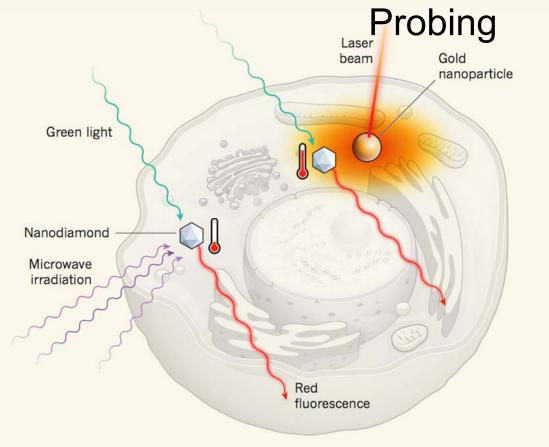


Texas A&M



has not been watered for 1 week.

Quantum Nanosystems and Quantum Optical



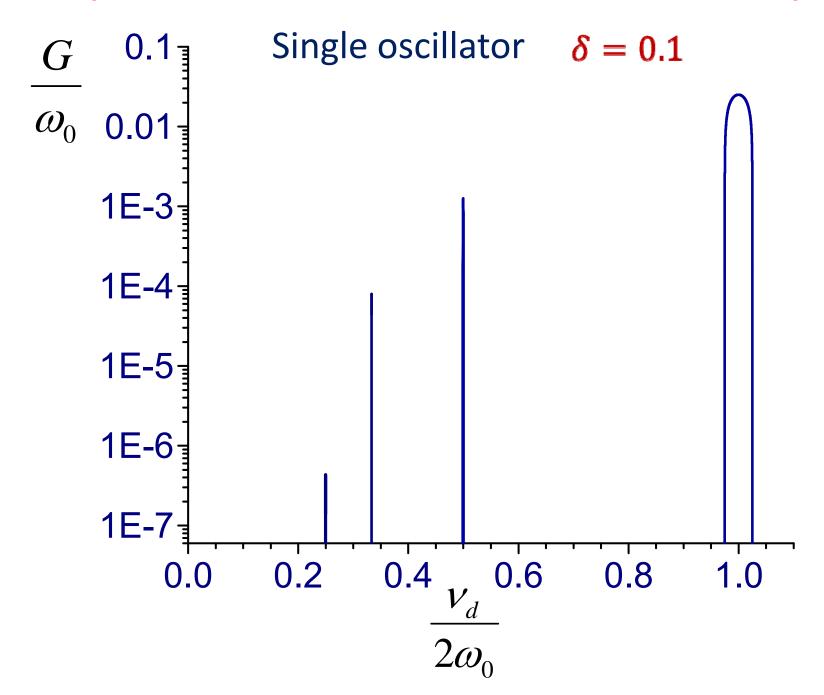


Mikhail Lukin, Harvard

Past: Demonstration of nanometer-scale thermometry in a living cell. Future: Use this technique to measure magnetic fields in cell and in the brain.

Kucsko, G.; Lukin, M.D., Aggie Laser Physics Ph.D. "Nanometer-scale thermometry in a living cell", Nature 500, 54 (2013).

Gain per unit time as a function of modulation frequency



Systems with many normal modes $\omega_1, \omega_2, \omega_3, ...$

$$v_d = \omega_2 + \omega_1 \leftarrow \text{Sum combination resonance}$$

$$v_d = \omega_2 - \omega_1 \leftarrow$$
 Difference combination resonance

Example: Coupled parametric oscillators

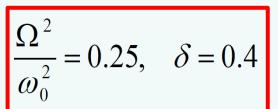
$$\ddot{x}_1 + \omega_0^2 x_1 - \Omega^2 x_2 = 0$$

$$\ddot{x}_2 + \omega_0^2 x_2 - \Omega^2 [1 + \delta \cos(v_d t)] x_1 = 0$$

For 1st order resonance:

$$G \propto \delta \cdot \Delta \omega$$

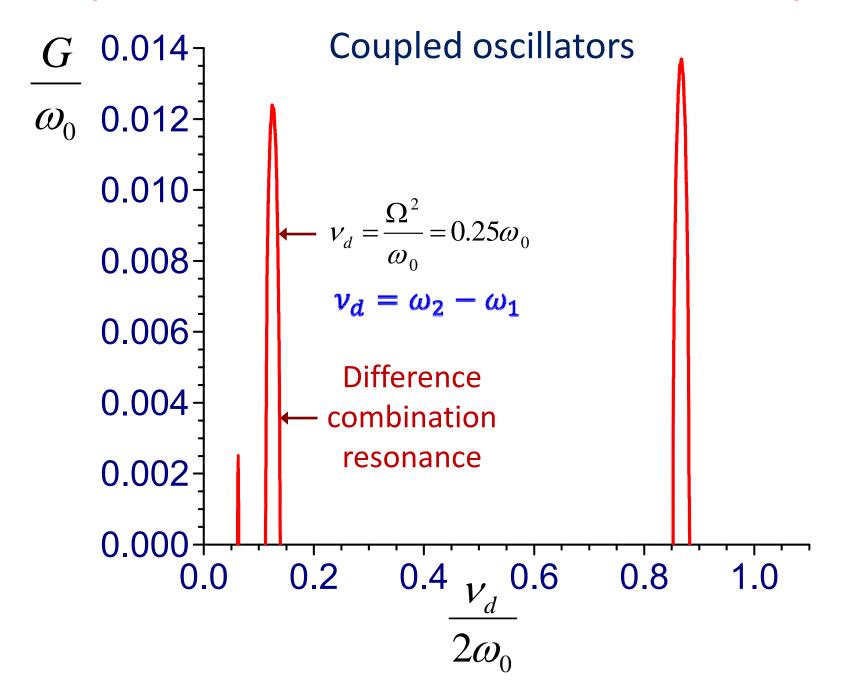
We solve equations numerically with



 $\Delta\omega$

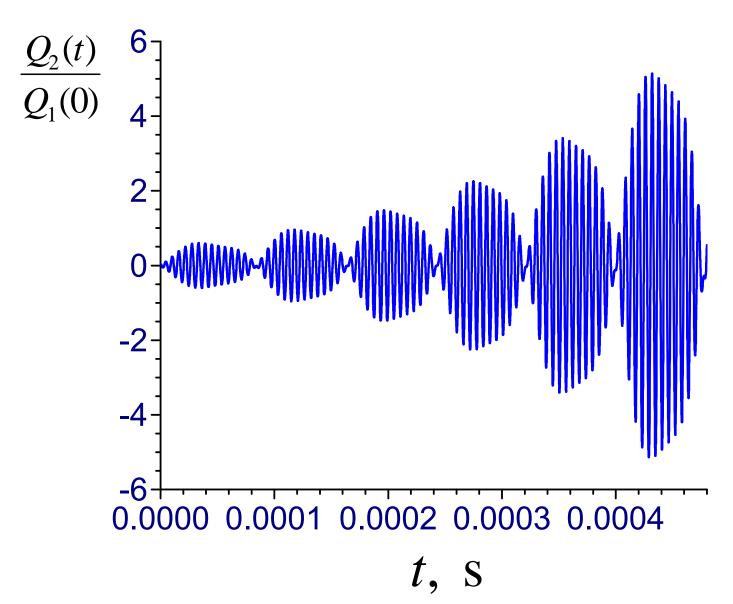
(1)

Gain per unit time as a function of modulation frequency

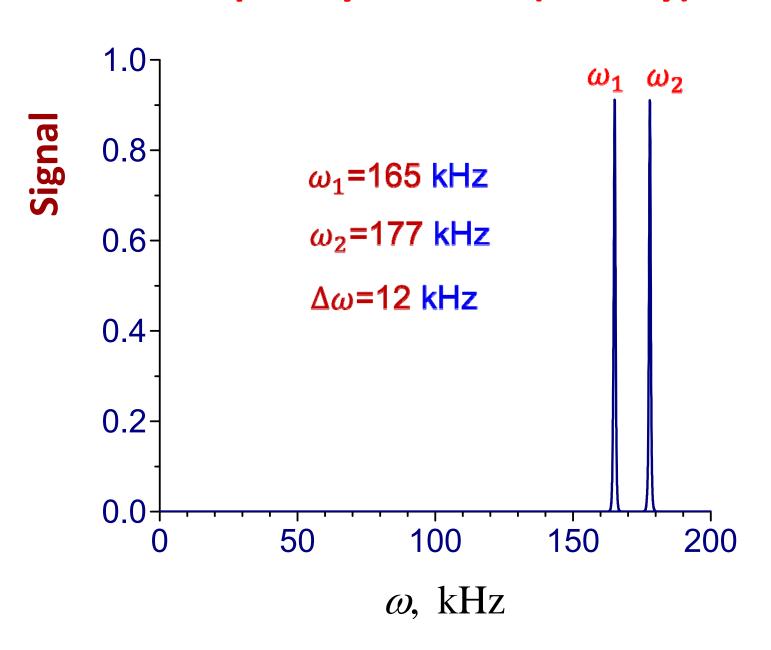


Oscillation of charge on capacitor C₂ (Theory)

With drive $\alpha V_0 = 0.1$, $\nu_d = 12$ kHz



In frequency domain (Theory)



Experimental demonstration of difference combination resonance

