

# Journal of Visualized Experiments

## Femtosecond Laser Filaments for Use in Sub-Diffraction-Limited Imaging and Remote Sensing

--Manuscript Draft--

Article Type:	Invited Methods Article - JoVE Produced Video
Manuscript Number:	JoVE58207R3
Full Title:	Femtosecond Laser Filaments for Use in Sub-Diffraction-Limited Imaging and Remote Sensing
Keywords:	Femtosecond filaments, remote sensing, ultrafast spectroscopy, sub-diffraction-limited imaging, nonlinear optics, laser-induced-breakdown spectroscopy
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**TITLE:**

Femtosecond Laser Filaments for Use in Sub-Diffraction-Limited Imaging and Remote Sensing

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**KEYWORDS:**

Femtosecond filaments, remote sensing, ultrafast spectroscopy, sub-diffraction-limited imaging, nonlinear optics, laser-induced-breakdown spectroscopy

**SUMMARY:**

High-intensity femtosecond pulses of laser light can undergo cycles of Kerr self-focusing and plasma defocusing, propagating an intense sub-millimeter-diameter beam over long distances. We describe a technique for generating and using these filaments to perform remote imaging and sensing beyond the classical diffraction limits of linear optics.

**ABSTRACT:**

Probing remote matter with laser light is a ubiquitous technique used in circumstances as diverse as laser-induced breakdown spectroscopy and barcode scanners. In classical optics, the intensity that can be brought to bear on a remote target is limited by the spot size of the laser at the distance of the target. This spot size has a lower bound determined by the diffraction limit of classical optics. However, amplified femtosecond laser pulses generate intensity sufficient to modify the refractive index of the ambient air and undergo self-focusing. This self-focusing effect leads to the generation of highly intense laser filaments which maintain their intensity and small sub-millimeter diameter size at distances well beyond the classical Rayleigh length. Such intensity provides the capability of remote scanning, imaging, sensing, and spectroscopy with enhanced spatial resolution. We describe a technique for generating filaments with a femtosecond regenerative chirped-pulse amplifier, and for using the resulting filament to conduct imaging and spectroscopic measurements at remote distances of at least several meters.

**INTRODUCTION:**

The spatial coherence and corresponding small divergence angle of laser beams have led to numerous applications in remote sensing, including chemically-sensitive measurements of the atmosphere<sup>1</sup>, range-finding<sup>2</sup>, and remote spectroscopy<sup>3</sup>. The same coherence properties allow very tight focusing of laser light that can deliver continuous focused intensities of billions of watts per square centimeter and pulsed intensities of  $10^{13}$  watts per square centimeter over a period of a few femtoseconds. Such extreme intensities are useful for numerous applications including examining the nonlinear optical properties of matter<sup>4</sup>, precision optical micromachining<sup>5</sup>, materials characterization through laser-induced-breakdown spectroscopy<sup>6</sup>, stimulated Raman spectroscopy, and trace chemical detection<sup>7</sup>.

However, the physical limitations of Gaussian beams set limits on the ability to apply these properties of extreme intensity and small divergence angle simultaneously. A laser beam focused to a small spot size will necessarily diverge with a greater angle. Classically, the beam divergence angle is given by  $\theta = \frac{2\lambda}{\pi w_0}$ , where  $\lambda$  is the wavelength and  $w_0$  is the radius of the beam waist. Since the divergence angle is set by the diameter of the laser beam and the focal length  $f$  of the focusing lens,  $\theta \approx \frac{d}{f}$ , and tight focusing is not possible at distances of many meters as  $f$  becomes large compared to  $d$ .

Workers in the field of amplified femtosecond pulses noticed that this limitation on intensity vs. range was violated for high-intensity femtosecond pulses, with burn marks smaller than the diffraction limit appearing on targets at large distance from the originating laser<sup>8</sup>. This was found to be due to Kerr-effect self-focusing. The refractive index of the air is modified in proportion to the intensity of the laser field, and when the laser has a Gaussian intensity profile, the resulting refractive intensity profile becomes functionally a lens<sup>4</sup>. The beam self-focuses as it propagates, resulting in a narrow and intense filament of less than 100  $\mu\text{m}$  radius whose small size is maintained by a dynamic balance among classical diffraction, Kerr self-focusing, and defocusing due to plasma generation<sup>9</sup>.

With femtosecond laser filaments, intensities on the order of  $10^{13}$  W/cm<sup>2</sup> can be delivered to targets at distances of many meters with commercially-available femtosecond chirped-pulse amplifiers. Thus, many experiments which previously required tight focusing conditions and targets very close to a lens of high numerical aperture can now be done at distance more typical of remote sensing applications. However, intensities much higher than this threshold are not easily possible with filamentation, as the beam tends to break up into multiple filaments where each individual filament is near the critical power for self-focusing<sup>9</sup>.

Numerous applications are possible. We present a protocol primarily applicable to imaging and spectroscopy of remote targets using a femtosecond laser filament scanned over the target surface. The experimental setup is shown in **Figure 1**.

## **Protocol:**

### **1. Creation of the Femtosecond Laser Filament**

1.1. As femtosecond filaments require the output of a Class 4 laser, wear appropriate eye protection rated for the particular laser system in use and establish a clear and well-defined beam line with an appropriate beam dump. Follow all standard laser safety procedures.

1.2. Begin with the output of a pulsed, amplified femtosecond laser whose instantaneous output power is greater than or equal to the critical power for self-focusing in air, about 3.2 GW for a Ti:Sapphire laser at 800 nm wavelength. Generate the amplified pulse in a commercial femtosecond laser amplifier system using manufacturer's protocol. In practice, pulse energy of about 1 mJ for an approximately 35 fs pulse is sufficient. Good results are obtained with pulse energy of 2-4 mJ.

1.3. Pass the laser beam through an iris that slightly clips the outer edges. It is observed to promote filament formation, since filament formation is known to be seeded by sharp gradients and inhomogeneity in the spatial intensity profile of the laser.

1.4. Pass the beam through the converging lens that has a focal length of approximately 200 cm or greater, so that the geometric focusing is not so great that self-focusing is overwhelmed by optical breakdown or diffraction. Slightly tilt the lens with respect to the direction of propagation, since additional anisotropy is known to help seed the self-focusing process.

1.5. Observe a filament at a location near the geometric focus of the lens. Diagnose filamentation by a diffuse (several-mm-sized) halo surrounding a bright (approximately 100- $\mu$ m-sized) core. The halo could be seen on a white paper and the bright cores usually flicker.

1.5.1. Additionally, observe a characteristic self-phase modulation process in the air, which produces bright, multi-colored conical emission rings that are visible beyond the filament. For lasers with energies which are several times the threshold for filamentation, multiple filaments are observed. These are visible as multiple bright spots in the conical emission pattern, and can be eliminated by attenuation before the iris.

## **2. Remote Scanning of the Target Surface**

2.1. Put a two-axis motorized translation stage capable of moving the sample in the direction transverse to the propagation of the laser beam on the table. Ensure that the laser beam is incident on the center of the stage. Bolt the stage on the table with screws. For laboratory purposes, it is generally easier to keep the laser beam fixed in space while scanning the target under the beam.

2.2. Place sand in a container (5 mm x 25.4 mm x 25.4 mm). The thickness of sand is around 2 mm.



2.2.1. Put the metals (copper, stainless steel, aluminum) on the top of sand (**Figure 3a**). Cover the metals with another 2 mm layer of sand (**Figure 3b**).

2.2.2. With the laser off, put the container in the center of the translation stage. Make sure that the center of container is at the location where filamentation is observed for step 1.1-1.5.

2.3. Set up the laser's computer control to fire a single shot when electronically commanded. Write a LabVIEW or a similar computer language to perform the control. For automated single-shot pulses, an external trigger is required.

2.3.1. Connect a trigger TTL pulse to the External Trigger port on the back of the laser control module with a BNC cable. Enable the external trigger option on the laser control module. The TTL pulse will now trigger the laser to fire a single shot.

2.4. Set up the appropriate sensor apparatus. Set up the entrance of the spectrometer pointing to the impact point.

2.4.1. Use a lens to couple the light from filamentation impact point into a spectrometer. Make sure that the distance between the lens and filamentation is about the focal length.

2.4.2. Connect the spectrometer with computer using USB cable. Use software to monitor the spectrum. Open the software and the spectrum, and then click the **Run** button.

2.4.3. Use the mouse to zoom in the range that is recorded in the experiment. Optimize the spectrometer position after seeing the signal on the screen.

2.4.4. For imaging measurements, replace the spectrometer with a photomultiplier tube or a CCD camera.

2.5. Write a program in LabVIEW or a similar computer language to perform a loop over the following steps: Fire a single shot from the laser; collect and save the resulting data; move the translation stage to the next coordinate point.

## REPRESENTATIVE RESULTS:

The resolution of the scanned images is limited optically only by the  $\sim 100\text{ }\mu\text{m}$ . Therefore, the translation stage motion should be of this order of magnitude or smaller for maximum resolution. However, this level of resolution is not necessary for all measurements. This protocol has been used for both imaging<sup>10</sup> and spectroscopic<sup>11</sup> measurements. **Figure 1** shows the experimental setup. The pulse is generated in an amplifier system. The pulse is 1 kHz, 50 fs, and centered at 800 nm. **Figure 2** compares a scan of a small Texas A&M logo target taken with a laser at the diffraction limit compared to a scan taken with a filament-forming beam. This experiment was performed using filaments in liquid water, but the results may be rescaled for air in remote sensing<sup>9</sup>. **Figure 3** shows spatially-resolved filament-induced breakdown

spectroscopy scans of metal objects of different composition buried approximately two millimeters below a layer of sand. The shapes and compositions of the metal objects are apparent. In general, filamentation provides a number of mechanisms for target effects. The initial pulse can provide information on the surface layer, while subsequent pulses can provide information on deeper portions of the material through ablation or mechanical removal of surface layers.

**Figure 1. The experimental setup.** The laser is 1 kHz, 50 fs, and centered at 800 nm. It is focused with a lens to reach the intensity ( $\sim 10^{13}$  W/cm<sup>2</sup>) to form laser filaments. The object is under sand and put on a translation stage. The scattered light is collected with a spectrometer.

**Figure 2. Sub-diffraction-limited Imaging.** Remote images generated by scanning a laser beam across a printed Texas A&M logo at a distance of several meters. a) Logo imaged with non-filamented beam. b) Logo imaged with filamented beam.

**Figure 3. The chemical map.** Spectrally and spatially resolved image of metal objects buried under sand. a) Objects above sand. b) Objects below  $2.3 \pm 0.3$  mm of sand. c) Image with material composition color-coded to metal spectral features. Composite image of the buried objects with aluminum (Al), copper (Cu), and stainless steel (SS) corresponding to the red, green, and cyan color components, respectively

## DISCUSSION:

The method presented above is a laboratory protocol for the use of high-intensity laser light delivered at classically intractable distances. Of the numerous possible applications of such light – CARS, FIBS, THz radiation, photoacoustics, superradiance, *etc.* – many applications can deliver point information about surface material properties. Femtosecond laser filaments with sub-classical-diffraction-limited spot size allows use of these techniques while scanning the surface on a point-by-point basis. This protocol is an ideal test bed for the development of such techniques.

The most critical aspect of the protocol is to generate the laser filamentation. To generate the stable laser filamentation, the critical laser intensity is a few  $10^{13}$  W/cm<sup>2</sup> and the clamped intensity is around  $1.4 \times 10^{14}$  W/cm<sup>2</sup> measured in experiment<sup>12</sup>. There is no laser filamentation when the intensity is either high or low. If the intensity is too high, the medium might be ionized strongly at the focal point and a laser induced break-down will happen. A bright spark instead of a laser filamentation will be observed. In that case, attenuate the power or use a lens with a longer focal length. Conversely, if the power is low (no plasma generation is observed), increase the power or use a lens with short focal length. Moreover, in either case, it is worthwhile to adjust the chirp to help to form a laser filamentation.

This scanning technique is generally better suited for laboratory use and proof-of-concept rather than field deployment since remote sensing in the field generally does not allow fine translation-stage control of the target under investigation. In those scenarios the same lab-developed laser techniques can be used, but the laser itself will have to be scanned through

more traditional beam steering methods such as changing the orientation of the laser apparatus itself.

The protocol could be relatively easily extended to involve experiments with multiple filaments, filament bundles, pump-probe experiments, standoff spectroscopy, waveguide, or numerous other possibilities. In each case, one of the major experimental hurdles is the alignment of the intersecting focal spots, but with this protocol, this need only be done once. The optical elements are fixed in place and the sample itself is the only object required to move. This can be done very precisely with a translation stage. Further modification of this protocol to achieve further control over the location of the filament formation distance, including filament formation at hundreds of meters from the laser, is possible in principle by careful control of the output laser pulse. Multi-filamentation will also form a waveguide during the propagation, which could help to deliver a light in free space.

Remote sensing is a broad subject that spans disciplines such as physics, chemistry, engineering, environmental science, *etc.* In the supplementary material, we propose additional remote sensing schemes including stand-off spectroscopy and superradiance in addition to filamentation.

#### **DISCLOSURE:**

No conflicts of interest declared.

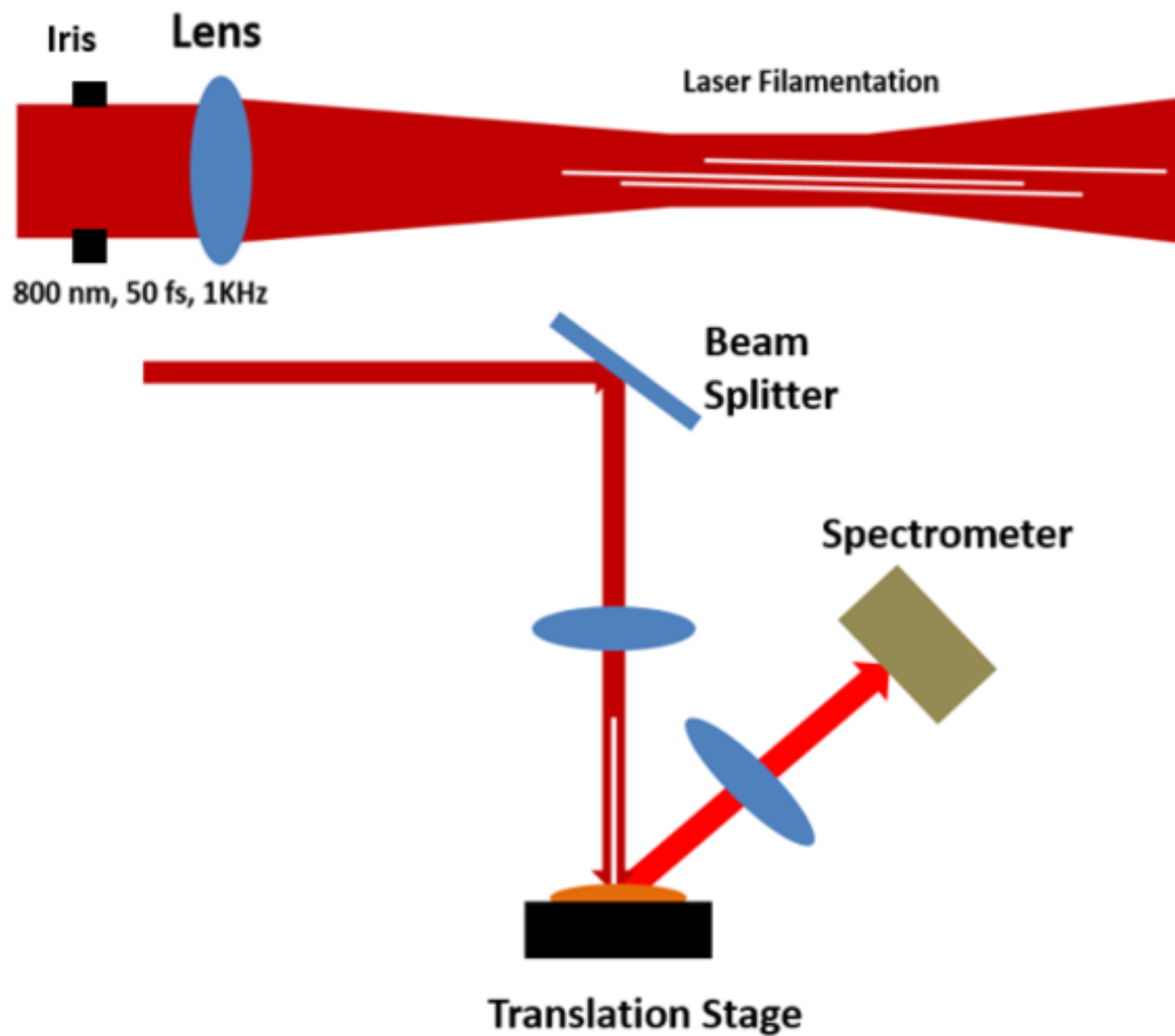
#### **ACKNOWLEDGEMENTS:**

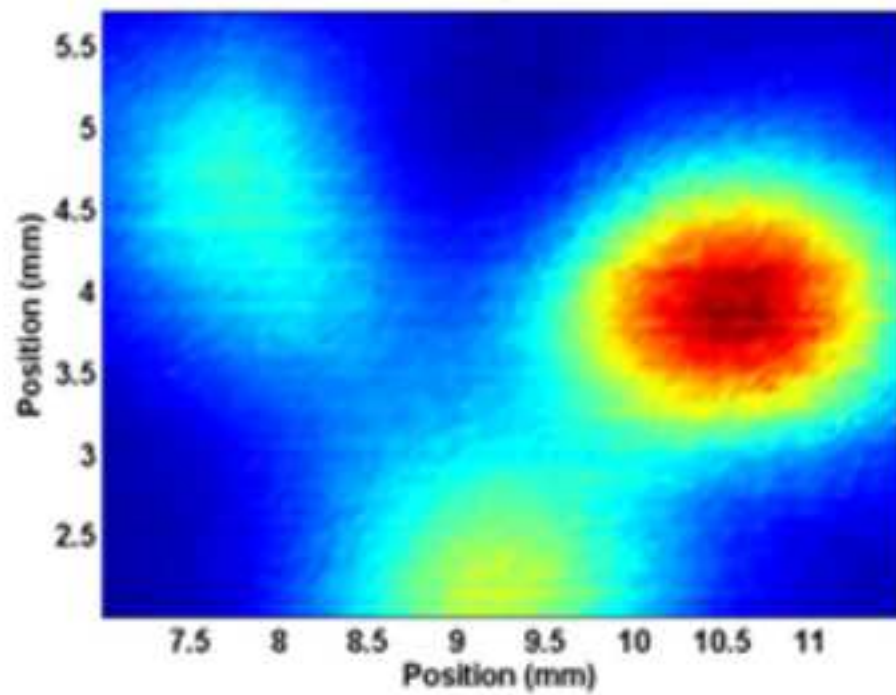
Funding provided by the Office of Naval Research, the Robert A. Welch Foundation (grant No. A-1547), and the SMART Fellowship.

#### **Reference**

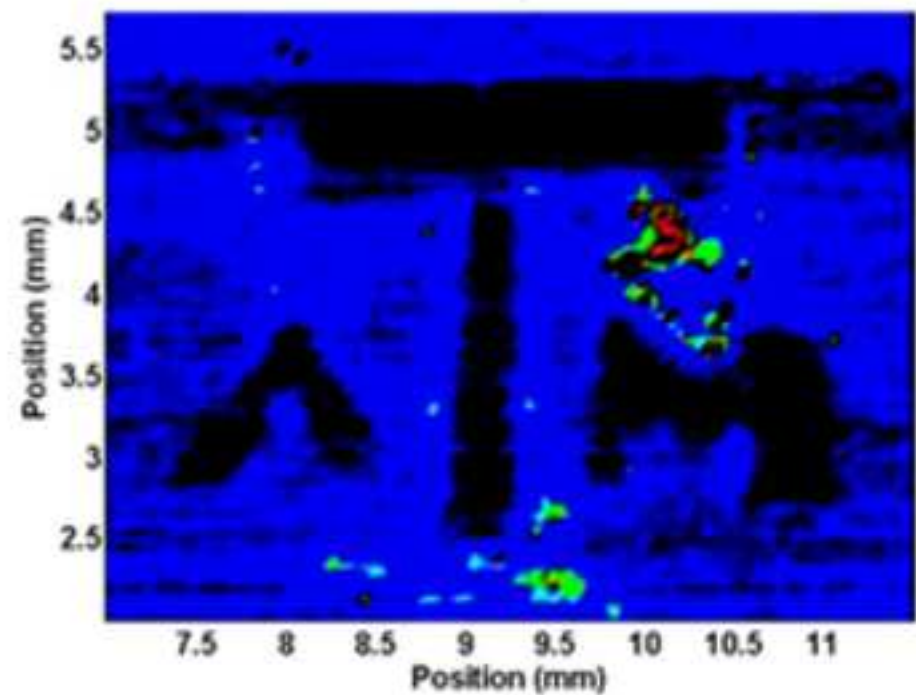
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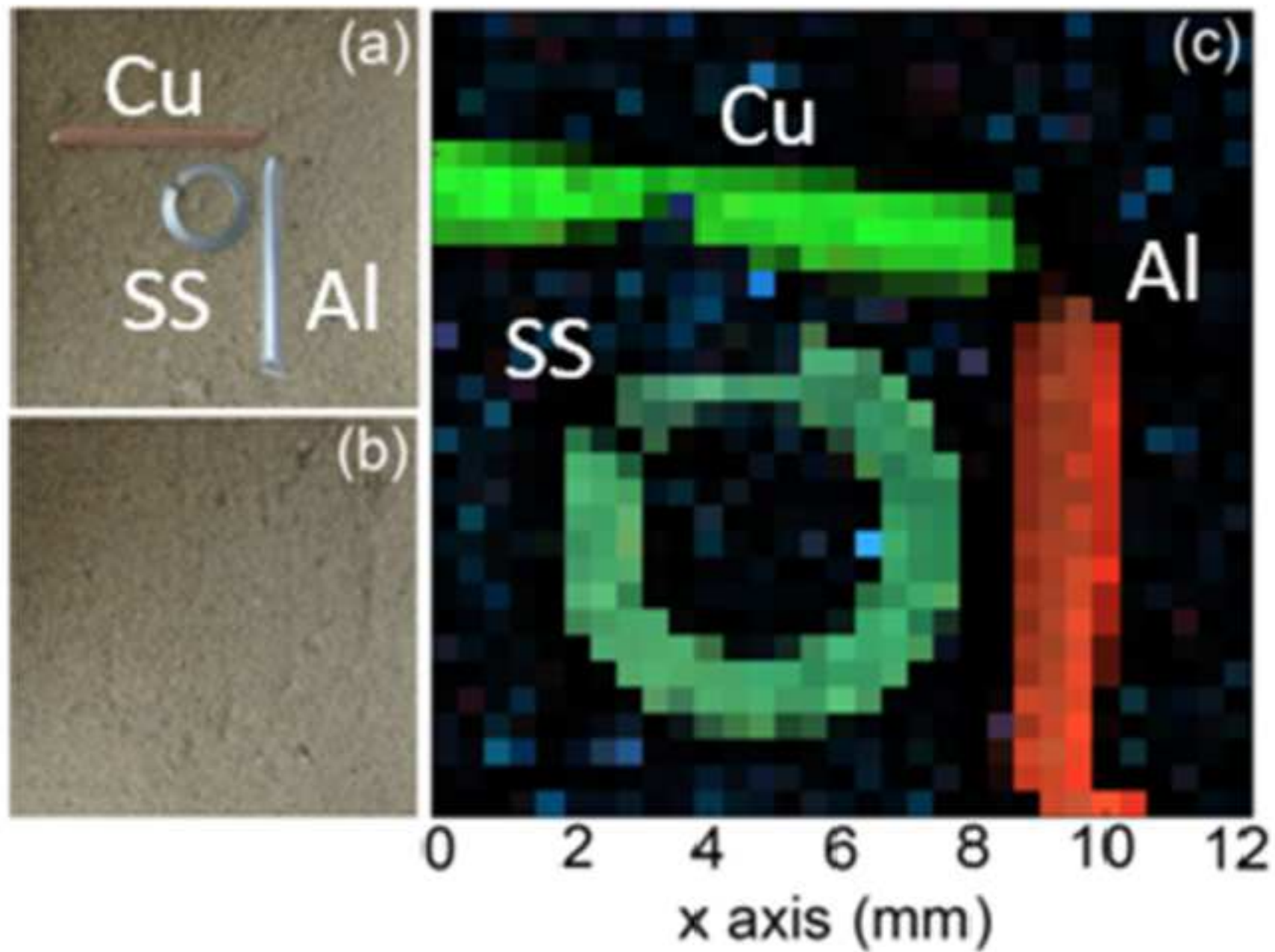




(a)



(b)





Name of Material/ Equipment	Company
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Femtosecond laser system	Coherent Co
IRIS	Thorlabs
Lens	Thorlabs
Mirrors	Thorlabs

Photodetector	Hamamatsu
Spectrometer	Ocean Optics
Translation Stage	Thorlabs



## Catalog Number

Legend Elite System

id25

LA1908-C

PF10-03-P01

H12694

OCEAN\_HDX\_VIS\_NIR

PT3-Z8

### **Comments/Description**

1 kHz system, fs system pulse energy 4 mJ  
Mounted Standard Iris, Ø25.0 mm Max Aperture, TR3 Post  
L=50 cm, Plano-Convex Lenses (AR Coating: 1050 - 1700 nm)  
Plano metallic mirror

Thermoelectric cooled NIR-PMT unit  
Spectrometer, high dynamic range, 350-950  
25 mm (0.98") Three-Axis Motorized Translation Stage, 1/4"-  
20 Taps



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### CORRESPONDING AUTHOR:

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Institution: Texas A&M University

Article Title: Femtosecond Laser Filaments for Use in Sub-Diffraction-Limited Imaging and remote sensing

Signature: [Signature] Date: 03/22/2018

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We appreciate the reviewers' comments. The following is our response.

#### Reviewer #1:

##### Manuscript Summary:

The manuscript Femtosecond Laser Filaments for Use in Sub-Diffraction-Limited Imaging and Remote Sensing is a very helpful article that covers experimental and theoretical details of remote sensing over a broad range of applications and methods. The article should be published after some substantial changes in the format.

##### Major Concerns:

The protocol described in lines 84 onward is very detailed but it most contain a cartoon of the experimental setup. There is a great contrast between the basic description of the protocol and the lack of graphic experimental detail and the theoretical description.

We thank the referee's comments. We add a new figure 1 to describe the experiment detail. We add sentences on page 3 before the protocol "...**The experimental setup is shown in Fig. 1.**" And sentences in the first paragraph on page 5 "... **Figure 1 shows the experimental setup. The pulse is generated in a amplifier system (Legend Elite, Coherent Co.). The pulse is 1KHz, 50 fs, and centered at 800 nm...**"

The theoretical description that follows after page 12 are of great scientific value as a compilation of work and should remain as part of the article. However, there seems to be a disconnect with the protocol described above. A link between Raman/Dicke at a distance and the protocol should be established early on. This disconnect is particularly strong after page 60 where a list of techniques seem to be listed but there is no "guide" to the reader on how the connection between experiments mentioned. While this connection can be done by a more specialized person, and might be good enough in that context, a connection between the experiments and the original protocol would greatly enhance the article.

The theoretical description is supplementary to our experimental protocol. It provides several proposals for remote sensing method using quantum optics. In the protocol we focus on the experiment of the generation and application of filamentation. We plan to have a special person to explain the connection between experiments and theory.

Finally, while the remote sensing aspect is present through out the discussion, the use of laser filaments is not clear and the Sub-Diffraction-Limited aspect seems to be completely absent from all discussions other than the nanosystems discussed in page 64.

We appreciate the comments. We note that the characteristic transverse sub-linear-diffraction width of filamentation phenomena makes it generally applicable for high-resolution sensing at longer distances and we have not belabored this point in the text. However, we have made a few revisions for additional clarity on this point. The modification is on page 5, the first paragraph of discussion is rewritten as "**The method presented above is a laboratory protocol for the use of high-intensity laser light delivered at classically intractable distances. Of the numerous possible applications of such light – CARS, FIBS, THz radiation, photoacoustics, superradiance, etc – many applications can deliver point information about surface material properties. Femtosecond laser filaments with sub-classical-diffraction-limited spot size allows use of these techniques while scanning the surface on a point-by-point basis. This protocol is an ideal test bed the development of such techniques.**"

#### Reviewer #2:

##### Manuscript Summary:

The protocol is described reasonably well and is suitable for publication after minor edits.

I 53-56: The authors suggest that extreme intensities of ultrafast intense laser pulses are useful for several application. In fact, none of the provided examples are appropriate. Extreme intensities are applicable to radiation generation, high-field science, and similar "intensity frontier" problems, whereas for the applications the authors listed the required intensities are many orders of magnitude smaller. In the context of filamentation, the typical intensity is  $10^{13}$  W/cm<sup>2</sup>, seven orders of magnitude lower than  $10^{20}$  W/cm<sup>2</sup> (which is still not the

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highest intensity demonstrated in laser experiments).

We appreciate the comments. The extreme intensities in our situation is on the order of  $10^{13}$  W/cm<sup>2</sup>. We correct it in the text. The applications we mention generally are not intensity-frontier phenomena, as they rely on relatively low-order nonlinearities. While filaments are versatile in a number of applications, we agree that they are inherently limited to the intensities given above. This is elaborated in the manuscript paragraph beginning "**With femtosecond laser filaments...**"

I 71: in addition to the two effects listed, the classical diffraction also contributes to the dynamic balance in filamentation

We thank the comments and we add the classical diffraction in the first paragraph on page 5. "radius whose small size is maintained by a dynamic balance **among classical diffraction**, Kerr self-focusing, and defocusing due to plasma generation [9].

"

I 127-131: it is not clear why the authors restrict the scope to single-shot measurement at one point, as it is not generally true that all material studied is always removed by a single shot

Typically, the single-shot regime is most useful for maximum speed of data collection as well as reducing the damage to the sample in those cases where this is a concern. For weaker signals, deeper interrogation of underlying material, and related cases it may be advantageous to operate in a multiple-shot regime. We have added language to this effect in the text. We modify sentence on page 5: "...**In general, filamentation provides a number of mechanisms for target effects. The initial pulse can provide information on the surface layer, while subsequent pulses can provide information on deeper portions of the material through ablation or mechanical removal of surface layers.**"

In Fig. 2, the colors should be explicitly labeled. What is the significance of (a) and (b) - does the result in (c) correspond to (a) or (b)?

Fig3. (a) shows the objects and Fig3. (b) shows that the objects are buried under sand. The result in Fig 3(c) is related with that of Fig 3(a). To make it clear, we update Fig.3 with label of metals.

### Reviewer #3:

#### Manuscript Summary:

This manuscript describes the use fs laser filaments for imaging and remote sensing. Though the examples given in the manuscript is not adequate for remote sensing application, the method described may be interesting to remote sensing and other communities.

#### Major Concerns:

The representative results discussed in the report are not adequate to highlights its use in image and remote sensing. For example, figure 2 provided LIBS scans of metal objects buried in sand. The mechanism behind the removal of sand may be shocks rather than ablation.

We appreciate the referee's comments. We update Fig.3 to enhance our results for the imaging and remote sensing.

Additionally, mechanical removal of coarse surface layering is a valid use case for remote sensing, e.g., in interrogating subsurface properties of materials with less pulse energy than is required for material removal via ablation.

#### Minor Concerns:

The authors commented about the applications of ultrahigh intense laser ( $\sim 10^{20}$  W/cm<sup>2</sup>) such as LIBS, Raman, material characterization etc. For all these applications, typically intensities  $\sim 10^{14}$  W/cm<sup>2</sup> are used. For example, see Applied Physics Reviews 5 (2), 021301 (2018)

#### Attachments area

We are grateful for the author to point it out. It should be around a few  $10^{13}$  W/cm<sup>2</sup> instead of  $10^{20}$  W/cm<sup>2</sup>. We correct it in the context.





1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.
2. Please provide at least 6 keywords or phrases (there are only 4 in your manuscript).

**We provide six keywords in the manuscript : Femtosecond filaments, remote sensing, ultrafast spectroscopy, sub-diffraction-limited imaging, nonlinear optics, laser-induced-breakdown spectroscopy**

3. For in-text referencing, please remove the brackets before and after the reference numbers.

**We remove the brackets before and after the reference numbers.**

4. Step 1.5: Please write this step in imperative tense as if telling someone how to do the technique.

**1.1. We rewrite this step in imperative tense. It is modified as: *Observe a filament at a location near the geometric focus of the lens. Diagnose filamentation by a diffuse (several-mm-sized) halo surrounding a bright (approximately 100- $\mu$ m-sized) core. Additionally, observe a characteristic self-phase modulation process in the air, which produces bright, multi-colored conical emission rings that are visible beyond the filament. For lasers with energies which are several times the threshold for filamentation, multiple filaments are observed. These are visible as multiple bright spots in the conical emission pattern, and can be eliminated by attenuation before the iris.***

5. 2.1: Please add more details to your protocol steps. Please ensure you answer the “how” question, i.e., how to set up the stage? Please explain the detail in more sub-steps.

**We re-write the step in detail. It is modified as: *Put a two-axis motorized translation stage capable of moving the sample in the directions transverse to the propagation of the laser beam on the table. Make sure that the laser beam is incident on the center of the stage. Bolt the stage on the table with screws. For laboratory purposes, it is generally easier to keep the laser beam fixed in space while scanning the target under the beam.***

6. 2.2: What sample? Please specify. How to prepare the sample?

**The sample is copper, stainless steel, aluminum in the experiment. We specify it in the manuscript as: *Place sand in a container (5 mm X 25.4 mm X 25.4 mm). The thickness of sand is around 2 mm. Put the metal copper, stainless steel, aluminum on the top of sand (Fig. 3(a)). Cover the metals with another 2 mm layer of sand (Fig. 3(b)). With the laser off, put the container in the center of the translation stage. Make sure that the center of container is at the location where filamentation is observed for step 1.1-1.5.***

7. 2.3: How to set up the laser’s computer control?

**We add instruction about how to setup the laser’s computer control. Please look at 2.3 : *Set up the laser’s computer control to fire a single shot when electronically commanded. For automated single-shot pulses, an external trigger is required. Connect a trigger TTL pulse to the External Trigger port on the back of the laser control module with a BNC cable. Enable the external trigger option on the laser control module. The TTL pulse will now trigger the laser to fire a single shot.***

8. 2.4: How to set up? Please add in more details so that the readers can replicate your protocol.

We rewrite 2.4 and give the detail about how to setup the data collection. It is modified as : *Set up the appropriate sensor apparatus. Set up the entrance of the spectrometer pointing to the impact point. Use a lens to couple the light from filamentation impact point into a spectrometer (Ocean Optics). The distance between the lens and filamentation is about the focal length. The spectrometer is connected with computer with USB cable. The software is SpectraSuite. Open the software and click on “run” button. Optimize the spectrometer position after you see the signal on the software. For imaging measurements, replace the spectrometer with a photomultiplier tube or a CCD camera.*

9. 2.5.1-2.5.3: These steps cannot be filmed unless there is a graphical user interface being used. For steps that are done using software, a step-wise description of software usage must be included in the step. Please mention what button is clicked on in the software, or which menu items need to be selected to perform the step.

We make our own program use Matlab so that there is no other softwares. We re-write step 2.5 to give the details. It is modified as: *Write a program in Labview or a similar computer language to perform a loop over the following steps: Fire a single shot from the laser; collect and save the resulting data; move the translation stage to the next coordinate point.*

10. Please revise the Discussion to explicitly cover the following in detail in 3-6 paragraphs with citations:

a) Critical steps within the protocol

**The critical step in the protocol is to generate laser filamentation. We modified the first paragraph on page 5:** *The most critical aspect of the protocol is to generate the laser filamentation. To generate the stable laser filamentation, the critical laser intensity is a few  $10^3 \text{ W/cm}^2$  and the clamped intensity is around  $1.4 \times 10^4 \text{ W/cm}^2$  measured in experiment [12]. There are no laser filamentation when the intensity is either high or low. If the intensity is too high, the medium might be ionized strongly at the focal point and a laser induced break-down will happen. A bright spark instead of a laser filamentation will be observed. In that case, attenuate the power or use a lens with a longer focal length. Conversely, if the power is low (no plasma generation is observed), increase the power or use a lens with short focal length. Moreover, in either case, it is worthwhile to adjust the chirp to help to form a laser filamentation.*

b) Any modifications and troubleshooting of the technique

**The most frequently troubleshooting of the technique is to generate filamentation. We modified the text to describe the troubleshooting. Please see it in the response 10 (a)**

c) Any limitations of the technique

**We have discussion of the limitation of the technique. Please look at the second paragraph on page 5:** *This scanning technique is generally better suited for laboratory use and proof-of-concept rather than field deployment in view of the*

***fact that remote sensing in the field generally does not allow fine translation-stage control of the target under investigation. In those scenarios the same lab-developed laser techniques can be used, but the laser itself will have to be scanned through more traditional beam steering methods such as changing the orientation of the laser apparatus itself.***

d) The significance with respect to existing methods

e) Any future applications of the technique

**We include (d) and (e) in the third paragraph on page 5. It is modified as: *The protocol could be relatively easily extended to involve experiments with multiple filaments, filament bundles, pump-probe experiments, standoff spectroscopy, waveguide, or numerous other possibilities. In each case one of the major experimental hurdles is the alignment of the intersecting focal spots, but with this protocol this need only be done once. The optical elements are fixed in place and the sample itself is the only object required to move, and this can be done very precisely with a translation stage. Further modification of this protocol to achieve further control over the location of the filament formation distance, including filament formation at hundreds of meters from the laser, is possible in principle by careful control of the output laser pulse. Multi-filamentation will also form a waveguide during the propagation, which could help to deliver a light in free space.***

11. Please do not use a table for References.

## Editorial comments:

The manuscript has been modified and the updated manuscript, **58207\_R2.docx**, is attached and located in your Editorial Manager account. **Please use the updated version to make your revisions.**

1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.
2. Step 1.2: How to generate the output of pulsed, amplified femtosecond laser? Please add more details.

**The pulse is generated in a commercial system (Legend Elite, Coherent Co.). We update this in the text: "...The amplified pulse is generated in a commercial femtosecond laser amplifier system (Legend Elite, Coherent Co.). In practice..."**

3. 1.5: How to diagnose? Please add more details. Please ensure that all text is written in imperative tense. Any text cannot be written in imperative tense should be added as a "Note."

**The diagnose is using white paper. It is colorful halo with bright spots flicker in the center. To make it clear, we modified the text as : "...a bright (approximately 100- $\mu$ m-sized) core. Note, the halo could be seen on a white paper and the bright cores usually flicker. Additionally, observe ..."**

4. 2.3: For steps that are done using software, a step-wise description of software usage must be included in the step. Please mention what button is clicked on in the software, or which menu items need to be selected to perform the step.

**There is not software used in this step. People have to make their own program to control the device. We add text to emphasize this "...Write a labview or a similar computer language to perform the control...."**

5. 2.4: Please ensure that all text is written in imperative tense. Any text cannot be written in imperative tense should be added as a "Note."

**We revise 2.4. It is modified as : "Set up the appropriate sensor apparatus. Set up the entrance of the spectrometer pointing to the impact point. Use a lens to couple the light from filamentation impact point into a spectrometer (Ocean Optics). Make sure that the distance between the lens and filamentation is about the focal length. Connect the spectrometer with**

**computer using USB cable. Use the software SpectraSuite to monitor the spectrum. Open the software and the spectrum click on “run” button. Use mouse to zoom in the range that is record in the experiment. Optimize the spectrometer position after see the signal on the screen. For imaging measurements, replace the spectrometer with a photomultiplier tube or a CCD camera”**

6. Please do not use a table for References.

7. Please bold the volume numbers for all references.

**We correct 6 and 7 in the text.**

# Using Quantum Coherence for Remote Sensing

**Marlan O. Scully**

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



**Texas A&M University**

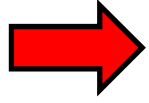
**Princeton University**



**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
- b) Coherence Brightened Air Laser

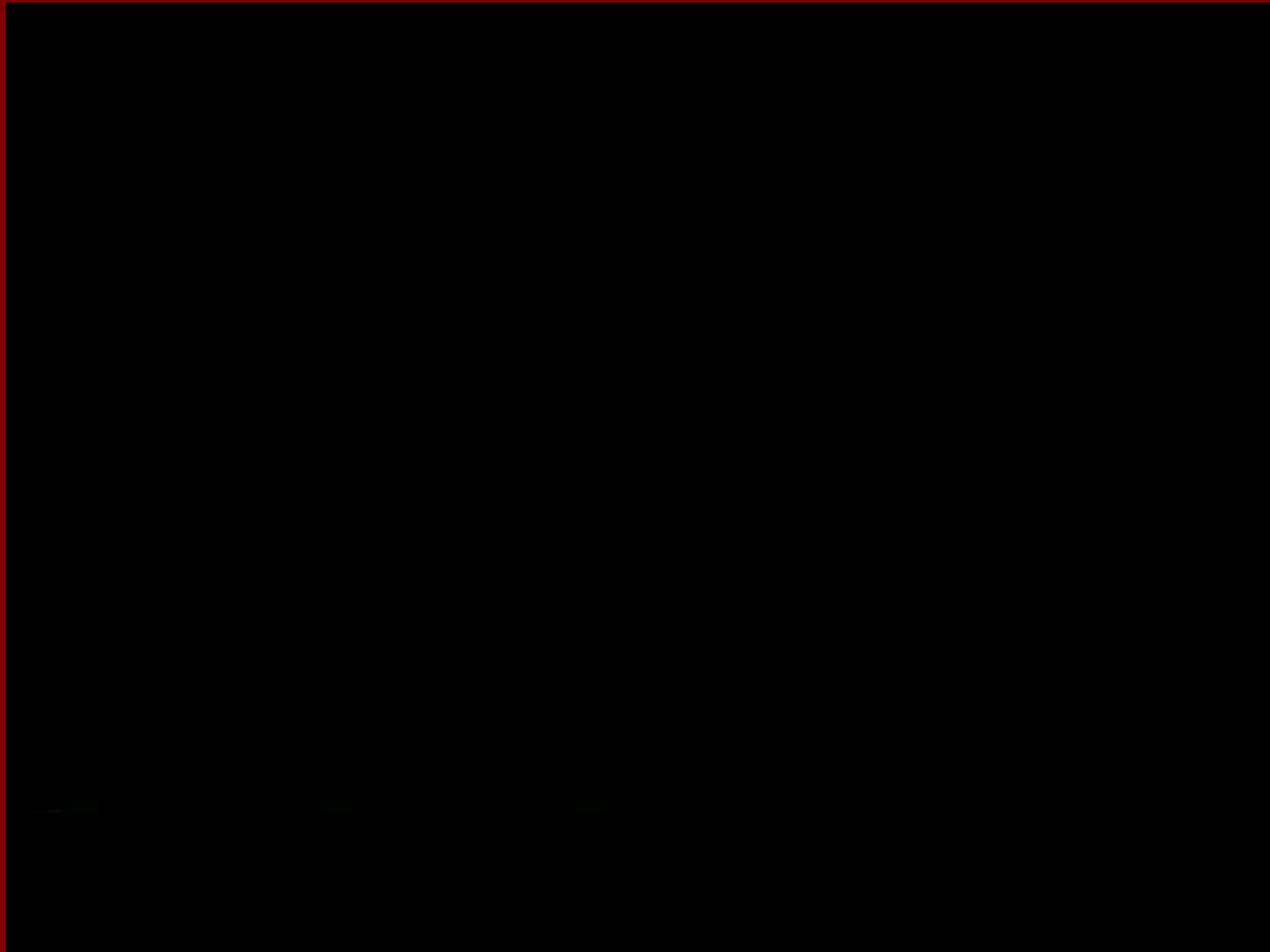
### 4) Backward Quantum Amplification by Superradiant Emission of Radiation

- a) Concept and Numerical Simulation
- b) Simple Gain Calculation

### 5) CARS in the Sky

### 6) Summary

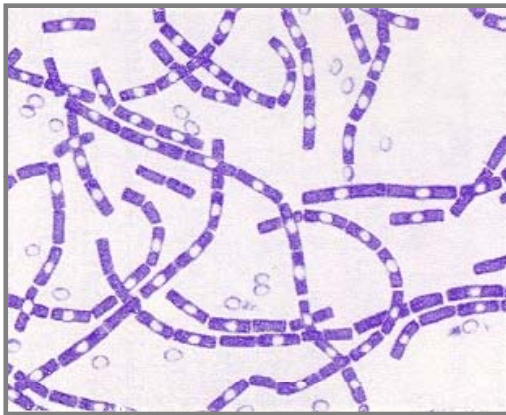
# NJ News Movie on Anthrax Detection





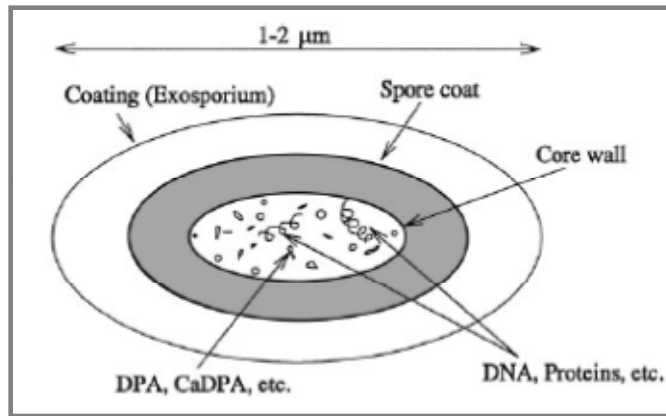
M.O. Scully et al., PNAS **99**, 10994 (2002)

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



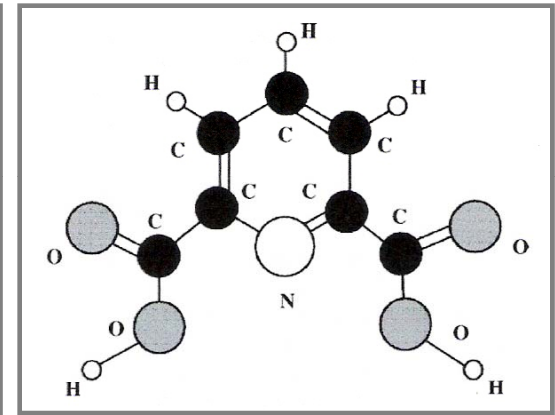
*Bacillus anthracis*

<http://textbookofbacteriology.net>



*Sketch of a spore*

M. O. Scully et al, PNAS, 2002



*Dipicolinic acid*

**Marker-molecule!**

**FAST CARS** – Femtosecond **A**daptive **S**pectroscopic **T**echnique  
for **C**oherent **A**nti-**S**tokes **R**aman **S**cattering

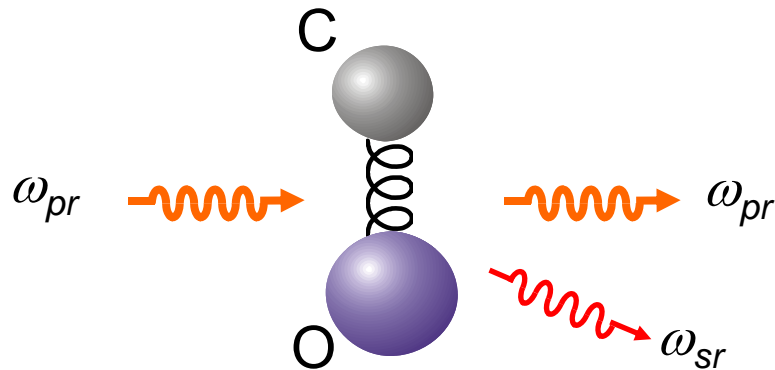
G. Beadie et al. (Washington, USA)

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)**



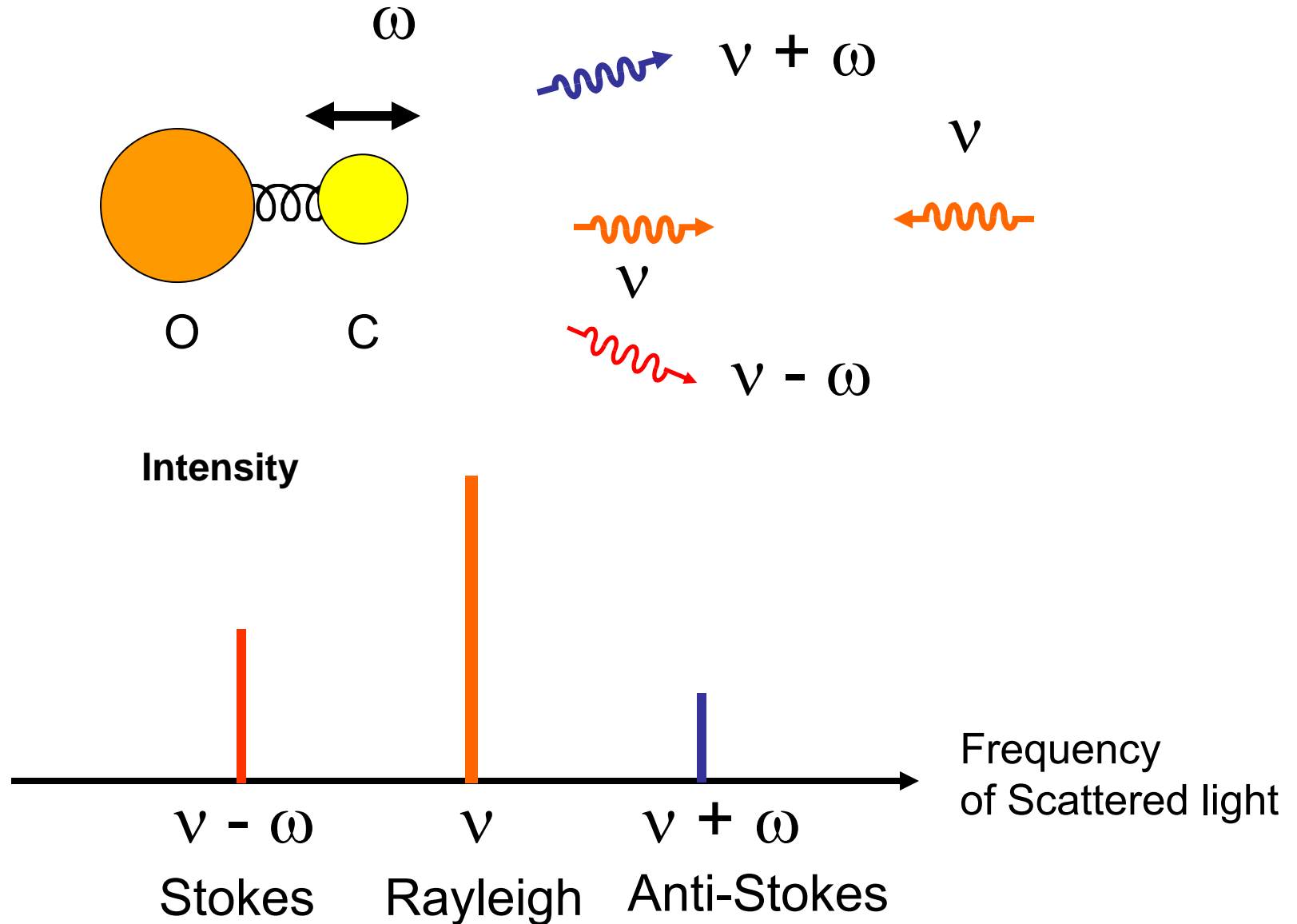
**AND**

**Chandrasekhara  
Venkata Raman  
(1888-1970)**

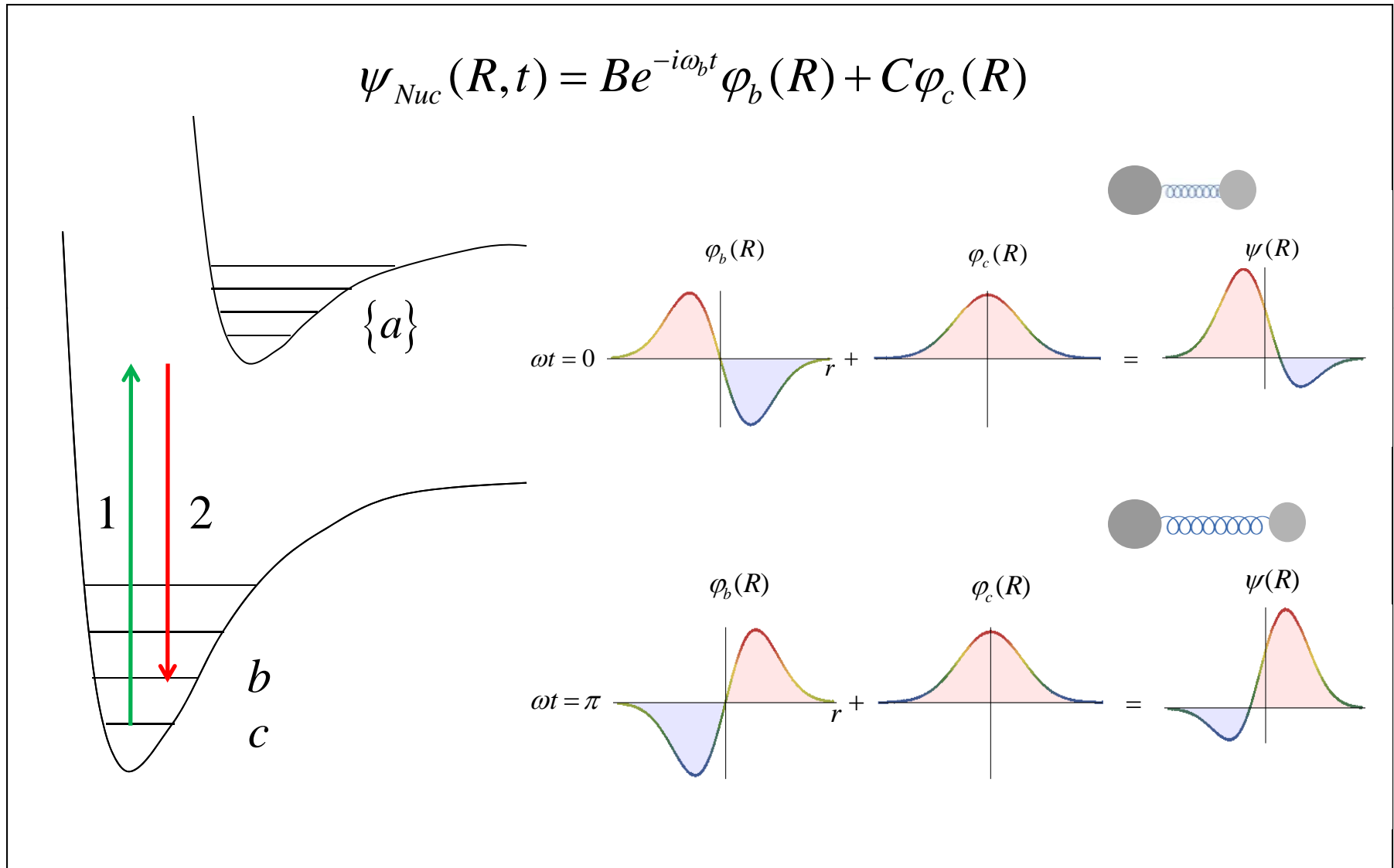
1930 Nobel  
in Physics



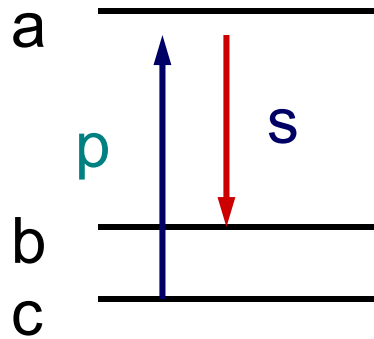
# Raman Scattering is like Reflection off Oscillating Mirror



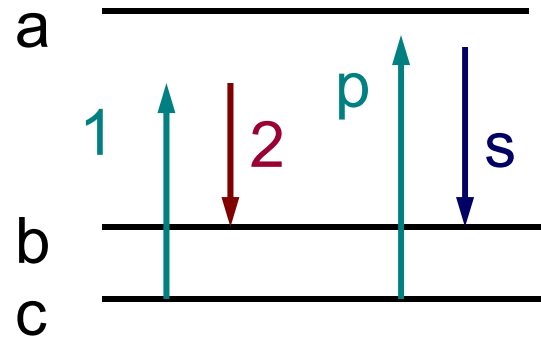
# Raman Coherence, $\rho_{bc}$ , implies Oscillation of Nuclear separation



## INCOHERENT



## COHERENT



## Raman Hamiltonian

$$H_{p,s} = \sum_j G(|b\rangle\langle c|)_j a_s^\dagger a_p e^{i(\vec{k}_p - \vec{k}_s) \cdot \vec{r}_j} + adj.$$

## Equation of motion for Stokes field

$$\dot{a}_s = -\frac{i}{\hbar} [H_{p,s}, a_s]$$

## Short Pulse

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

# Number of Stokes photons

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

$$\langle n_s(\tau) \rangle \cong \left\{ \sum_i \rho_{cc}^i + \sum_{i \neq j} \rho_{bc}^i \rho_{cb}^j e^{i[\vec{k}_1 + \vec{k}_p - \vec{k}_2 - \vec{k}_s] \cdot (\vec{r}_j - \vec{r}_i)} \right\} \left( \frac{G\tau}{\hbar} \right)^2 \langle n_p \rangle$$

Spontaneous

Incoherent

One Atom

Cooperative

Coherent

N Atom

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

Incoherent Raman

Coherent Raman

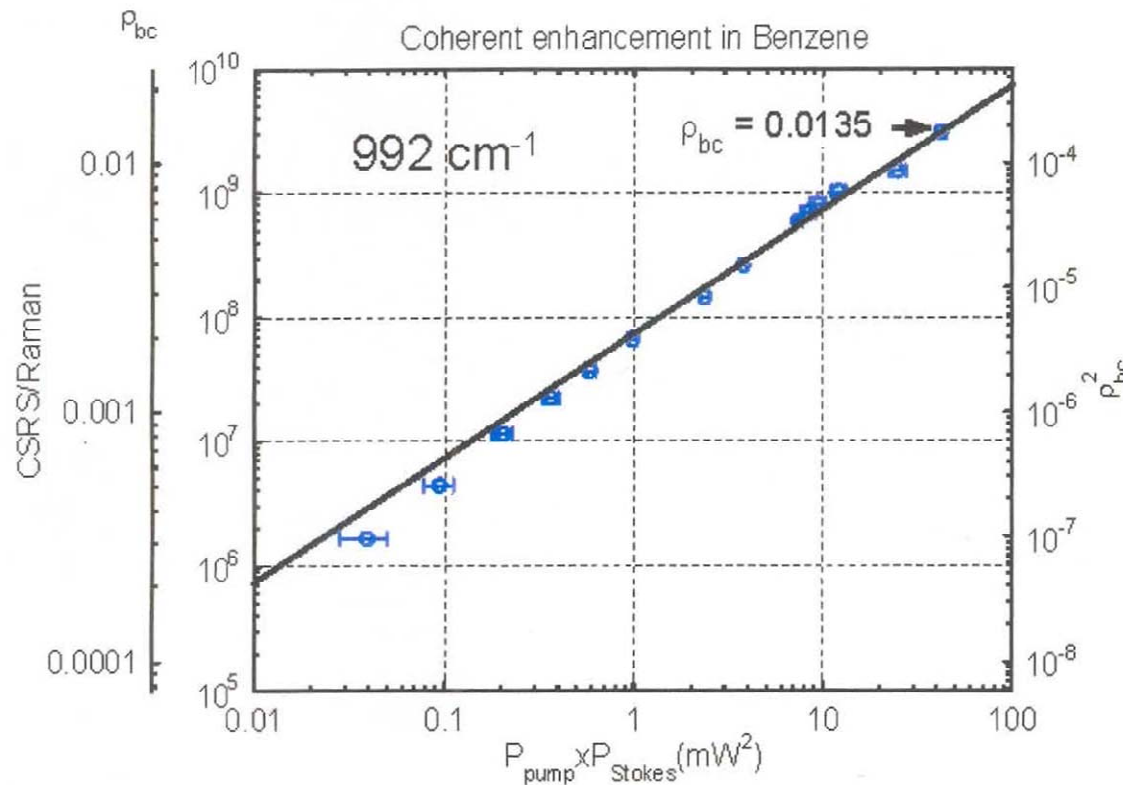
**Bottom Line**

$$\boxed{\frac{\langle n_s \rangle_{coh}}{\langle n_s \rangle_{incoh}} \cong \frac{N}{V} \lambda^2 R \frac{|\rho_{bc}|^2}{\rho_{cc}}}$$

**Cooperative Spontaneous Emission**



# Raman “amplification” via coherence: Experimental coherence measurement



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

$$\rho_{bc}, \text{ experimental} = 0.0135$$

$$\rho_{bc}, \text{ theoretical} = 0.018$$

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



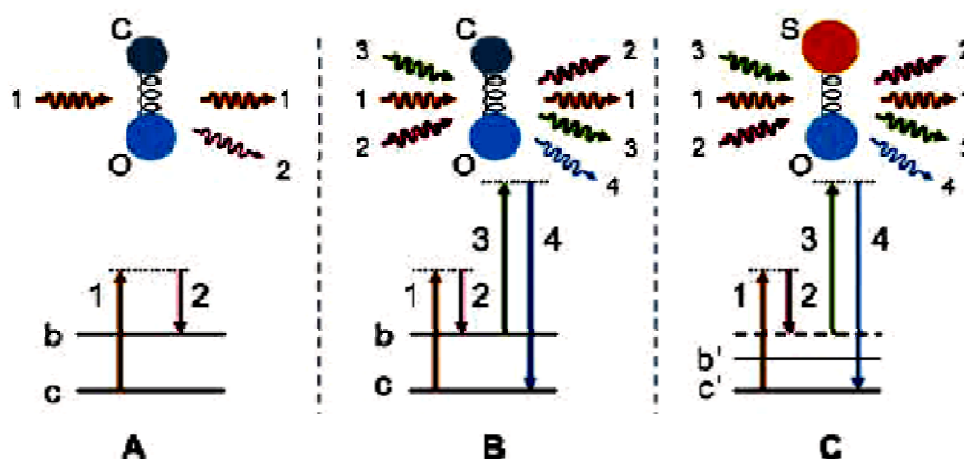
## Optimizing the Laser-Pulse Configuration for Coherent Raman Spectroscopy

Dmitry Pestov,<sup>1\*</sup> Robert K. Murawski,<sup>1,2</sup> Gombojav O. Ariunbold,<sup>1</sup> Xi Wang,<sup>1</sup> Miaochan Zhi,<sup>1</sup> Alexei V. Sokolov,<sup>1</sup> Vladimir A. Sautenkov,<sup>1</sup> Yuri V. Rostovtsev,<sup>1,2</sup> Arthur Dogariu,<sup>2</sup> Yu Huang,<sup>2</sup> Marlan O. Scully<sup>1,2</sup>

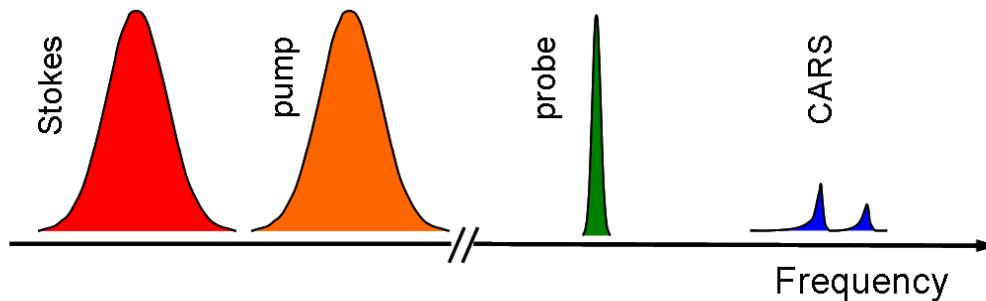
**Fig. 1.** Level diagram and schematic of different scattering processes on simple molecules. In this example, CO is a target molecule and SO is a background molecule.

**(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.

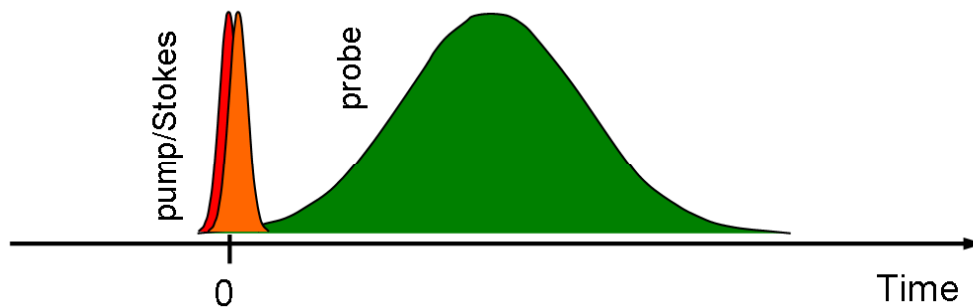
$c$ , the ground state of the CO molecule;  $b$ , the target vibrational state of the CO molecule;  $c'$ , the ground state of the background molecule;  $b'$ , an off-resonant vibrational state of the background molecule.



# Optimal CARS Technique



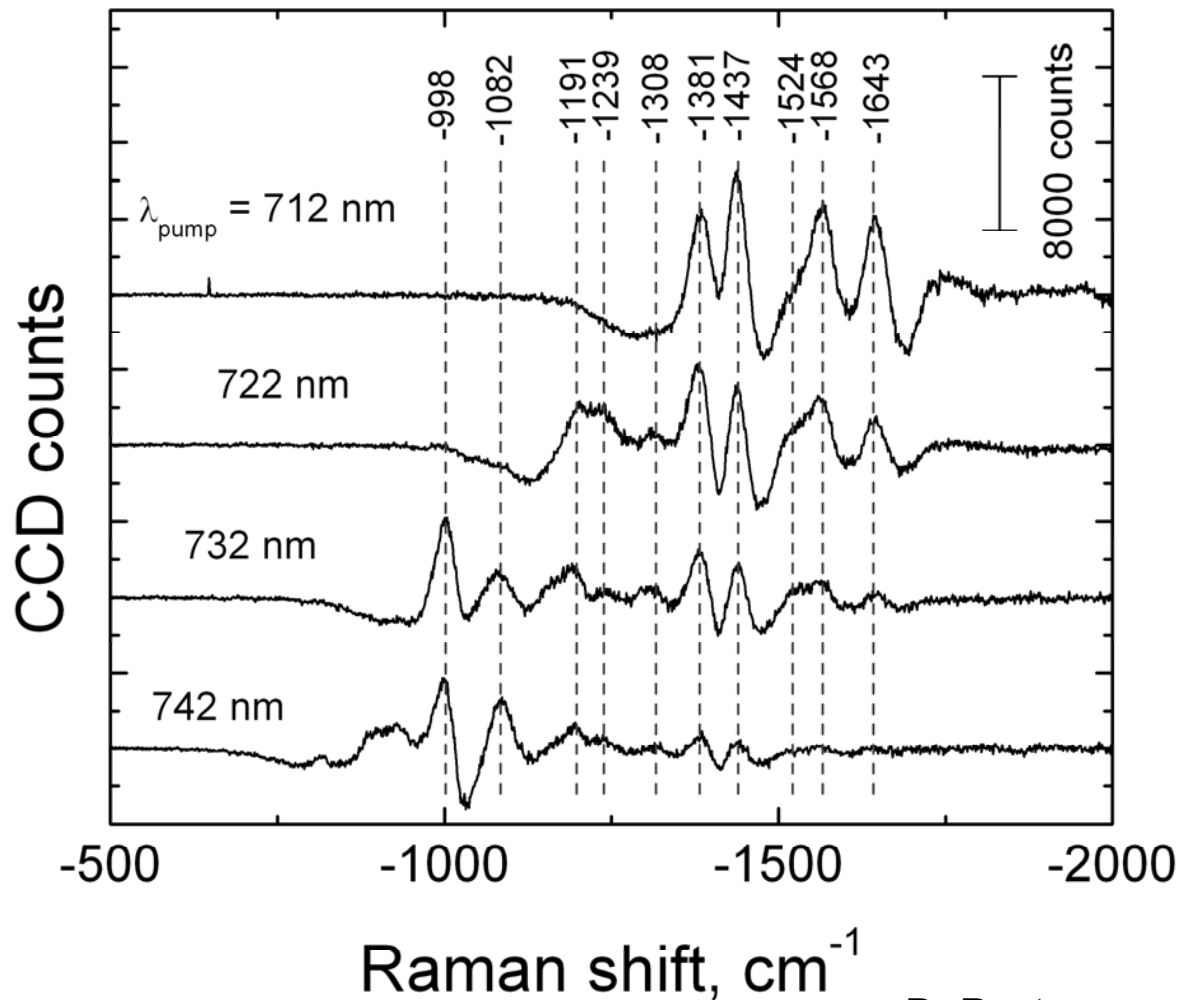
😊 Efficient broadband excitation and frequency-resolved probing, multi-channel detection => comprehensive species-specific information.



😊 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  $\mu\text{J}/\text{pulse}$

Stokes  
803 nm,  
3.9  $\mu\text{J}/\text{pulse}$

Probe  
577.9 nm,  
FWHM~0.7 nm  
~0.5  $\mu\text{J}/\text{pulse}$

Int. time is 2 min.

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

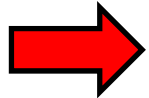
# Take-Home Message

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

## 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

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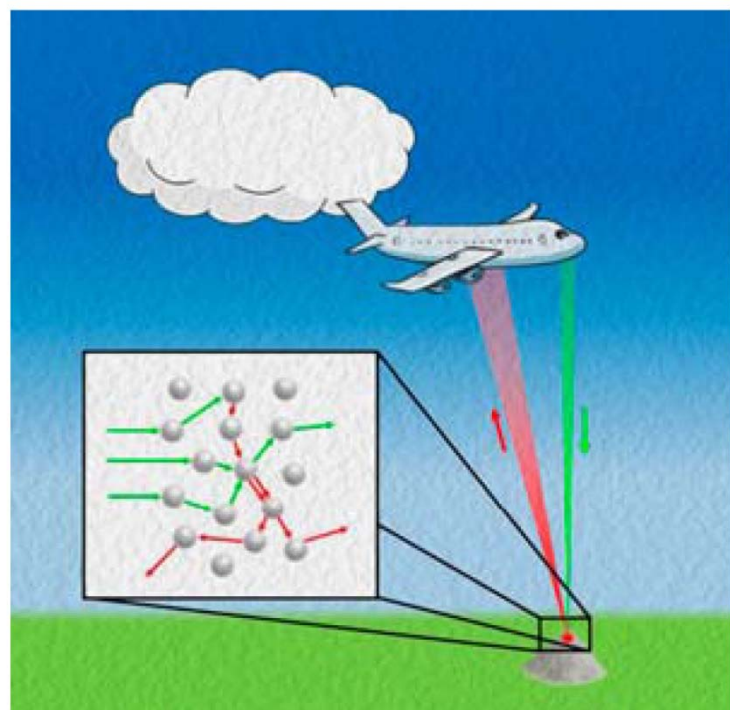
### 5) CARS in the Sky

### 6) Summary



# Photon pinball identifies chemicals from afar

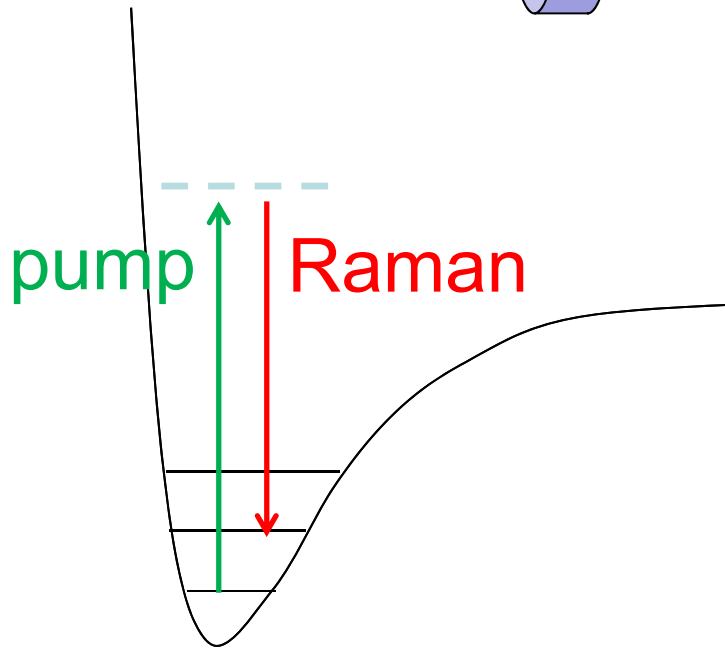
13 August 2014 [Andy Extnance](#)



Photons bouncing through a powder can create a 'random laser', also amplifying Raman scattering © NAS

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.

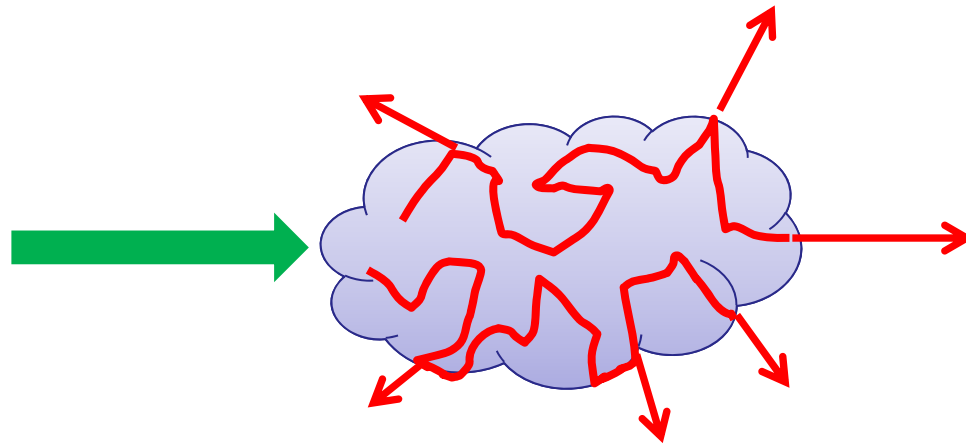
# What is a Raman laser?



Raman medium converts green pump photons to red Raman laser. Cavity contains red light.



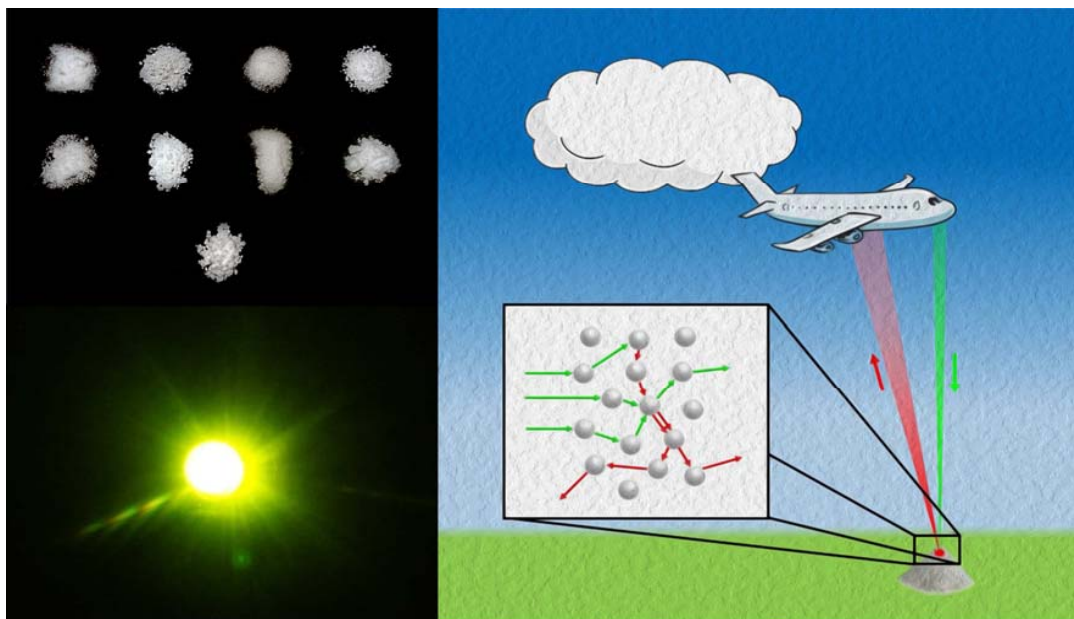
# What is a **random Raman laser**?



no  
cavity

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.

PNAS, 111 (34), 12320-12324 (2014)

# 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.

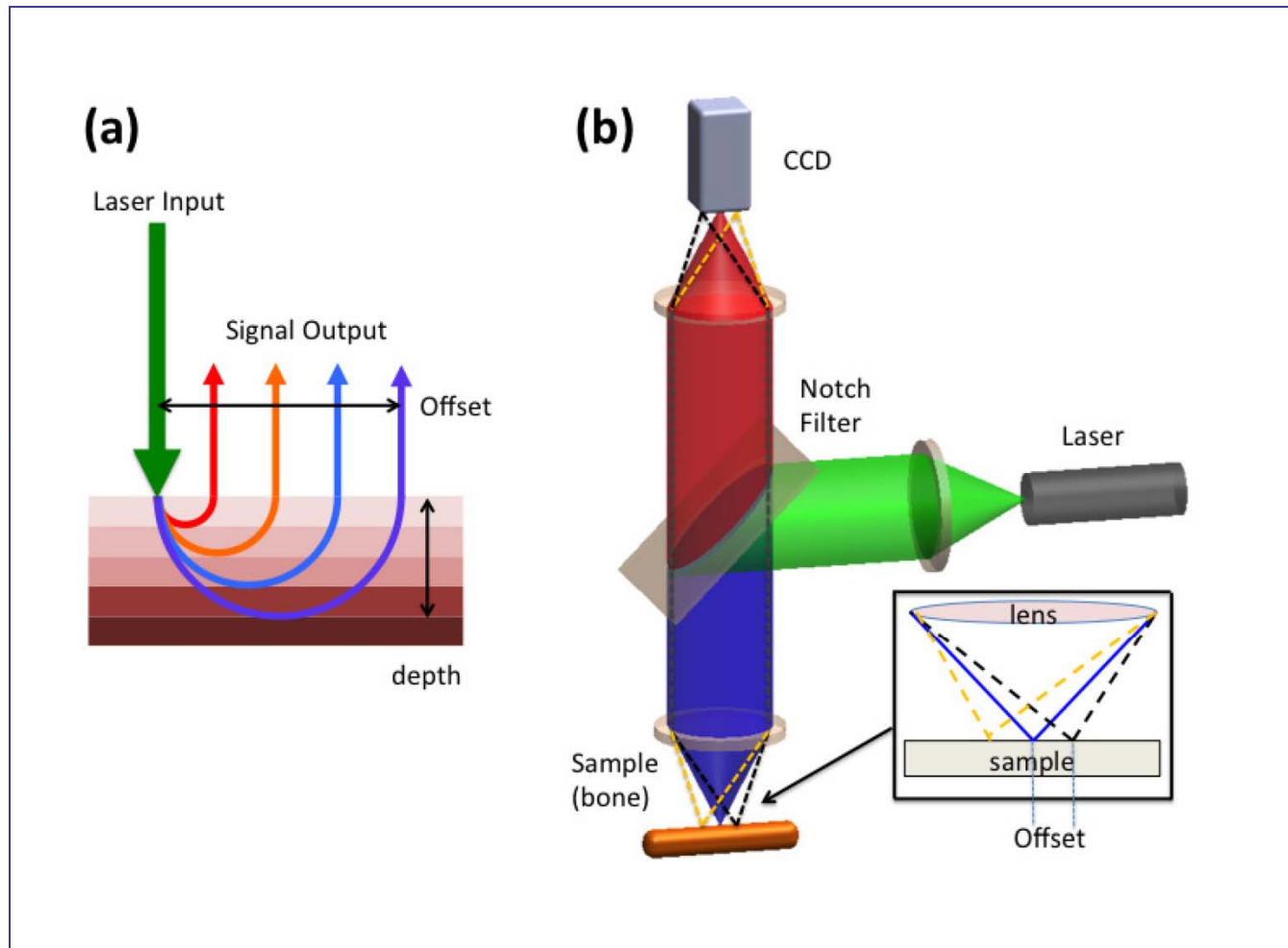


FIG. 1: Conceptual figure for (a): spatially offset Raman spectroscopy (SORS); (b): schematic diagram of the experimental microscope Raman detection setup.

## RAMAN AT RANGE

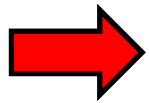
### 1) Anthrax Detection via FAST CARS

- 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

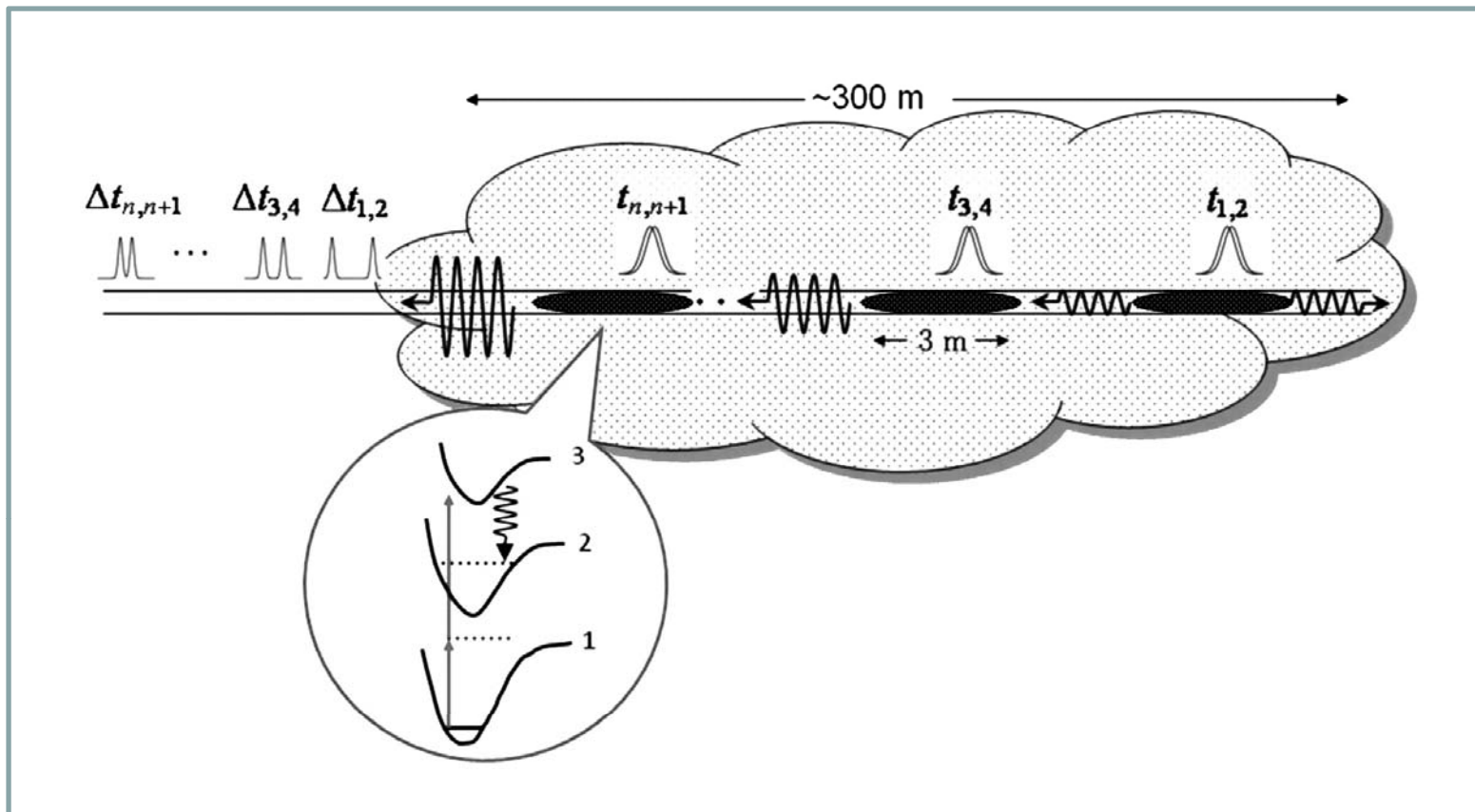
### 6) Summary

# 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  $O_2$ .



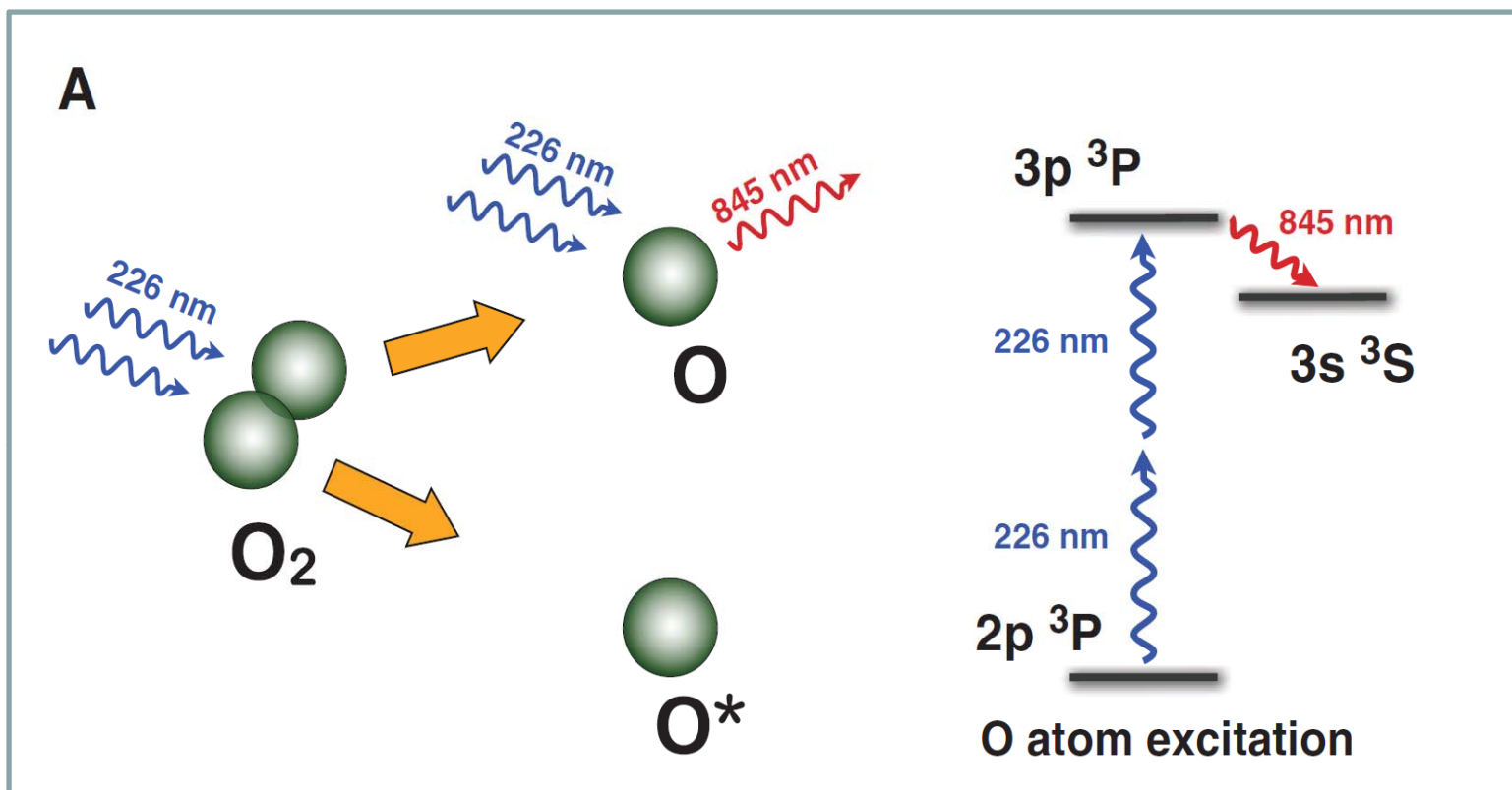
**Fig. 1.** SOS. Multiple pairs of pulses are generated such that the spacing between pulses in each pair is decreasing. The second pulse in each pair has a higher velocity because of atmospheric dispersion. The first pair of pulses overlaps near the back of the cloud, creating a small region of gain. 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.”

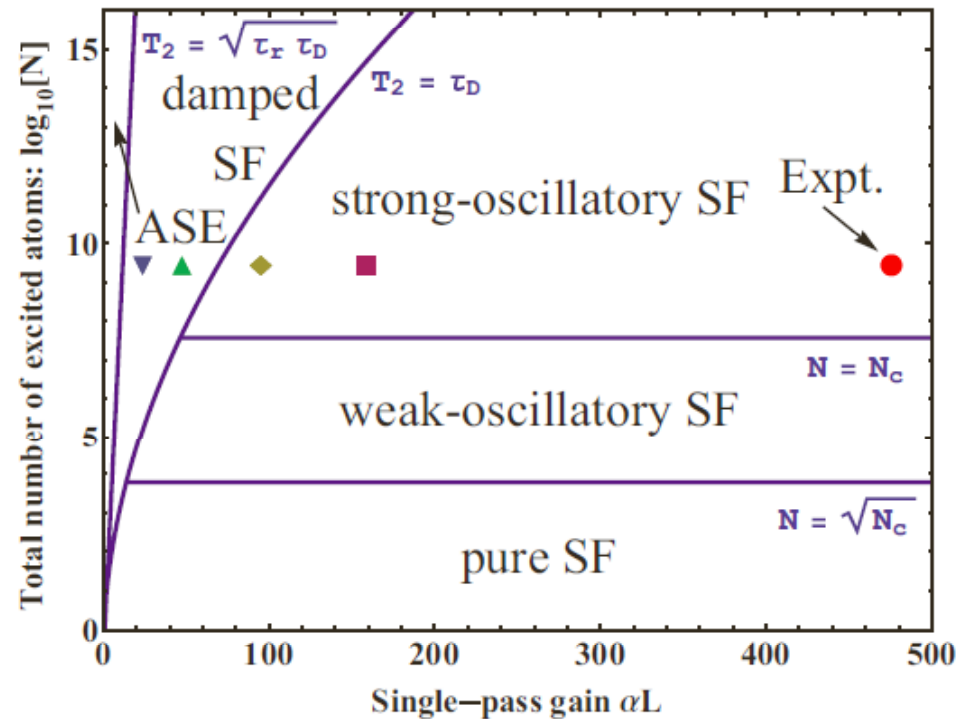


FIG. 1. (Color online) Regimes of  $N$ -atom cooperative spontaneous emission (adapted from Boyd et al.).

\*PNAS, **109**(38),15185-15190 (2012)

## RAMAN AT RANGE

### 1) Anthrax Detection via FAST CARS

- a) Motivation
- b) Coherent vs. Spontaneous Raman (Boyd, Shen, Welch)

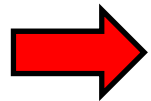
### 2) Random Raman

- 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



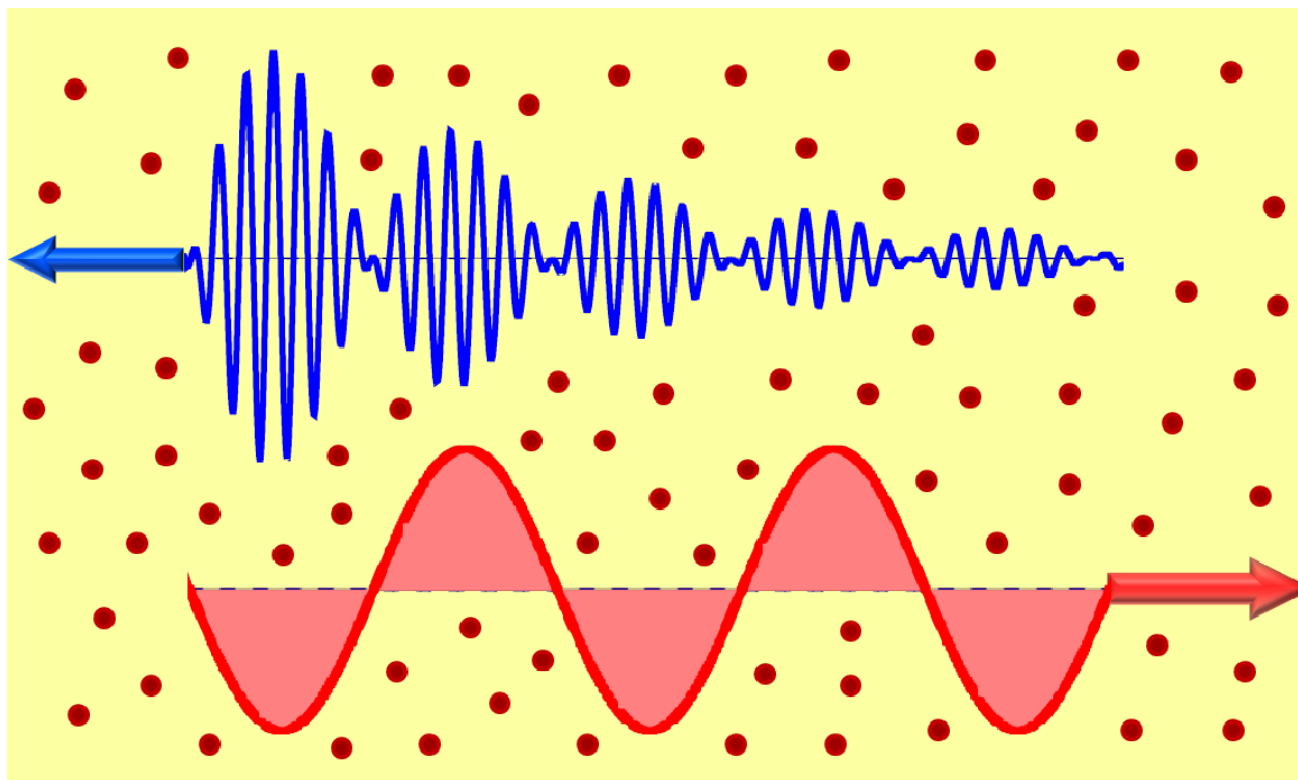
### 4) Backward Quantum Amplification by Superradiant Emission of Radiation

- a) Concept and Numerical Simulation
- b) Simple Gain Calculation

### 5) CARS in the Sky

### 6) Summary

# Quantum Amplification by Superradiant Emission of Radiation (QASER)



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

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# Quantum Amplification by Superradiant Emission of Radiation

Anatoly A. Svidzinsky,<sup>1,2</sup> Luqi Yuan,<sup>1,2</sup> and Marlan O. Scully<sup>1,2,3</sup>

<sup>1</sup>*Texas A&M University, College Station, Texas 77843, USA*

<sup>2</sup>*Princeton University, Princeton, New Jersey 08544, USA*

<sup>3</sup>*Baylor 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!

## PHYSICS

# The Super of Superradiance

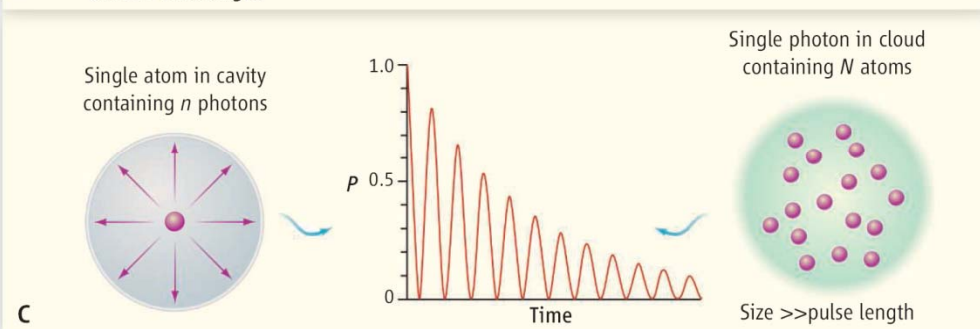
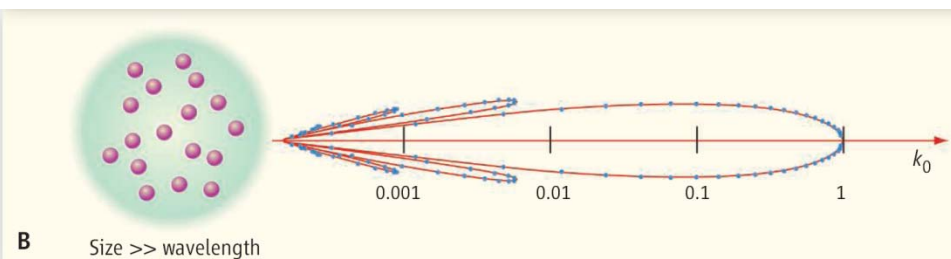
Marlan O. Scully<sup>1,2</sup> and Anatoly A. Svidzinsky<sup>1</sup>

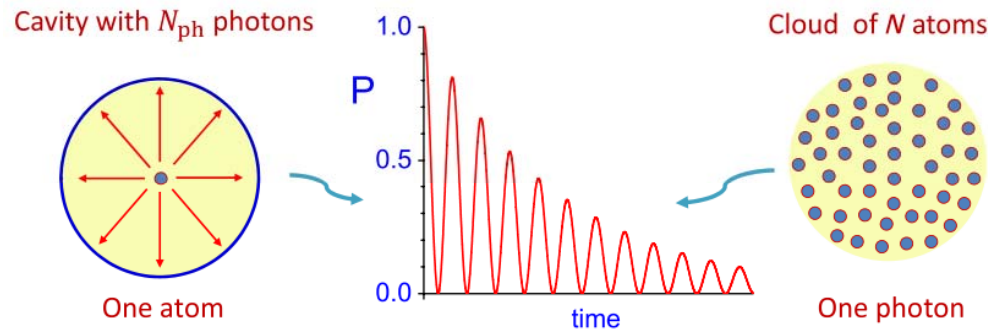
Cooperative single-photon emission from an atom ensemble will provide insights into quantum electrodynamics and applications in quantum communication.

## Timed Dicke state

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

A





	Probability of atomic excitation	Key variables V and N
Single atom maser	$\sin^2 \frac{\wp}{\hbar} \sqrt{\frac{\hbar \nu}{\epsilon_0 V}} \sqrt{N_{ph}} t$	$V$ = photonic cavity volume $N_{ph}$ = number of photons in cavity
Single photon superradiance	$\sin^2 \frac{\wp}{\hbar} \sqrt{\frac{\hbar \nu}{\epsilon_0 V}} \sqrt{N_{at}} t$	$V$ = atomic cloud volume $N_{at}$ = number of atoms in cloud

Can rewrite the collective superradiant oscillation frequency as:

$$\frac{\wp}{\sqrt{\hbar \epsilon_0}} \sqrt{\frac{\hbar \nu}{\epsilon_0 V}} \sqrt{N_{at}} = \sqrt{\frac{3}{4\pi} \lambda^2 \gamma c \frac{N_{at}}{V}} \equiv \Omega_a$$

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

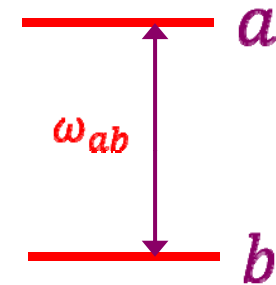
$$\left( \frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) \Omega_s = i \Omega_a^2 \rho_{ab}$$

# Physics of QASER

Electromagnetic field and atoms are two coupled oscillators

Maxwell's equation for electric field

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



Schrodinger equation for atoms

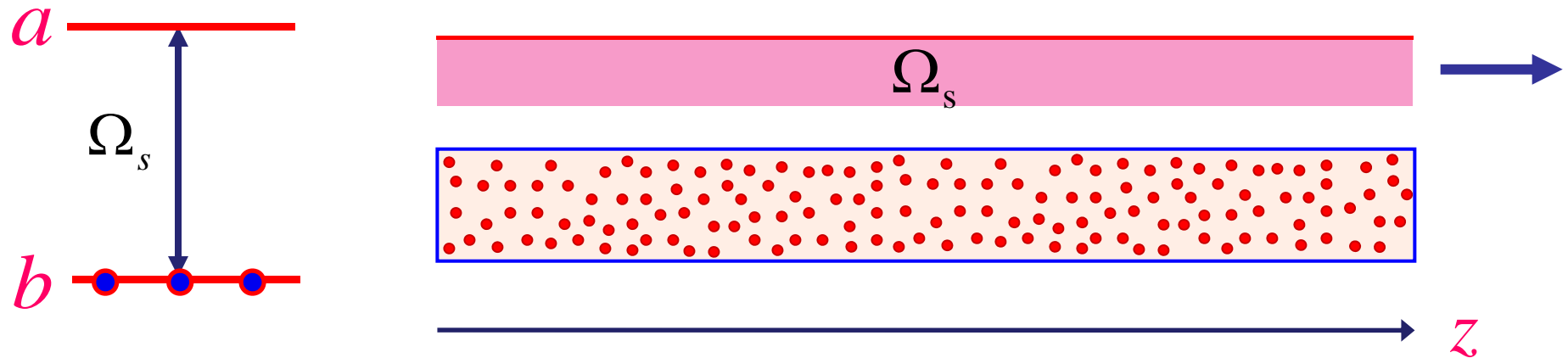
$$\frac{\partial \rho_{ab}}{\partial t} + i \omega_{ab} \rho_{ab} = \boxed{i \Omega (\rho_{bb} - \rho_{aa})}$$

Coupling between oscillators is nonlinear

Driving of atoms modulates coupling strength



# Light amplification with no population in excited state



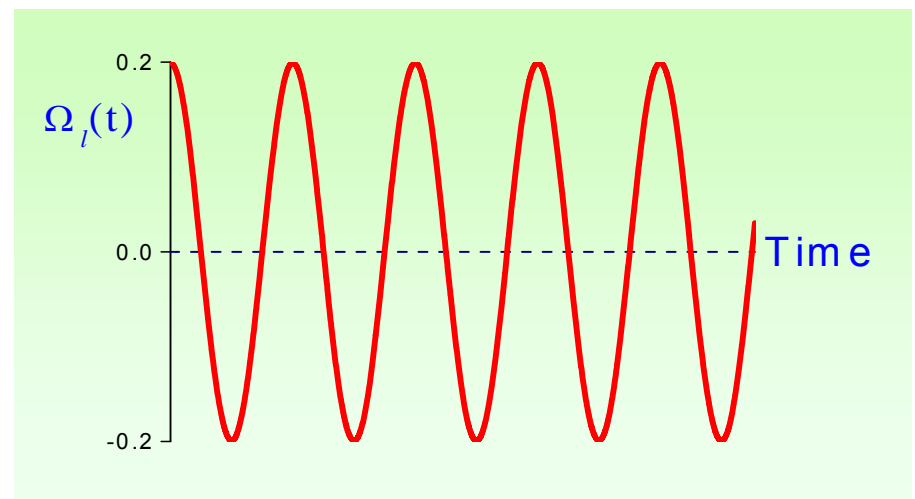
For uniform lasing field we obtain:

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

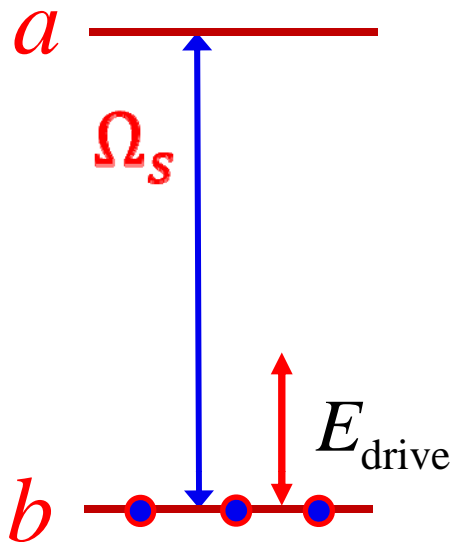
$$\Omega_a = \sqrt{\frac{3cN\lambda^2\gamma}{8\pi}}$$

Laser field oscillates with collective frequency:

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



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**

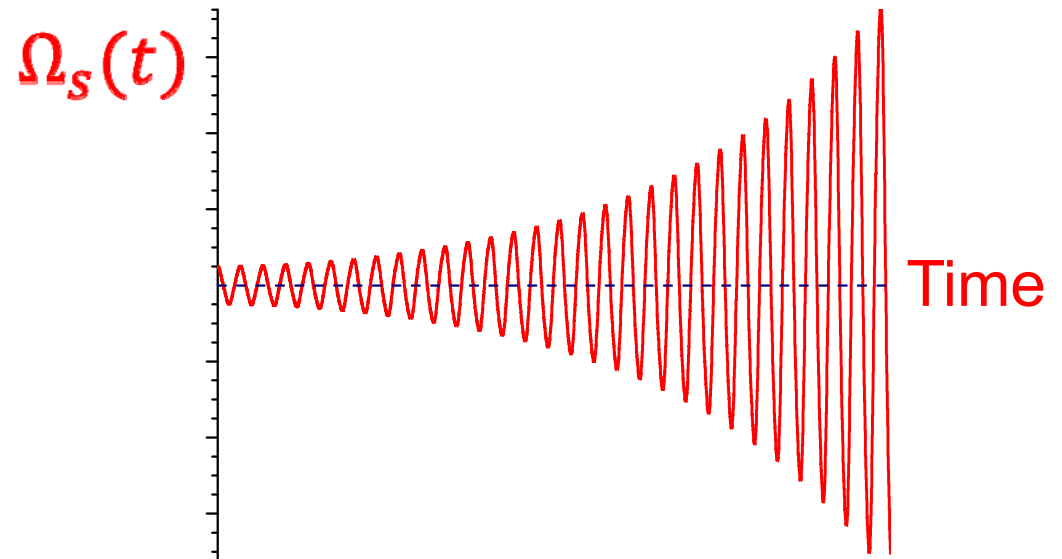
$$\frac{\partial^2 \Omega_s}{\partial t^2} + \Omega_a^2 [1 - \delta \cos(2\nu_d t)] \Omega_s = 0 \quad \delta \ll 1$$

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

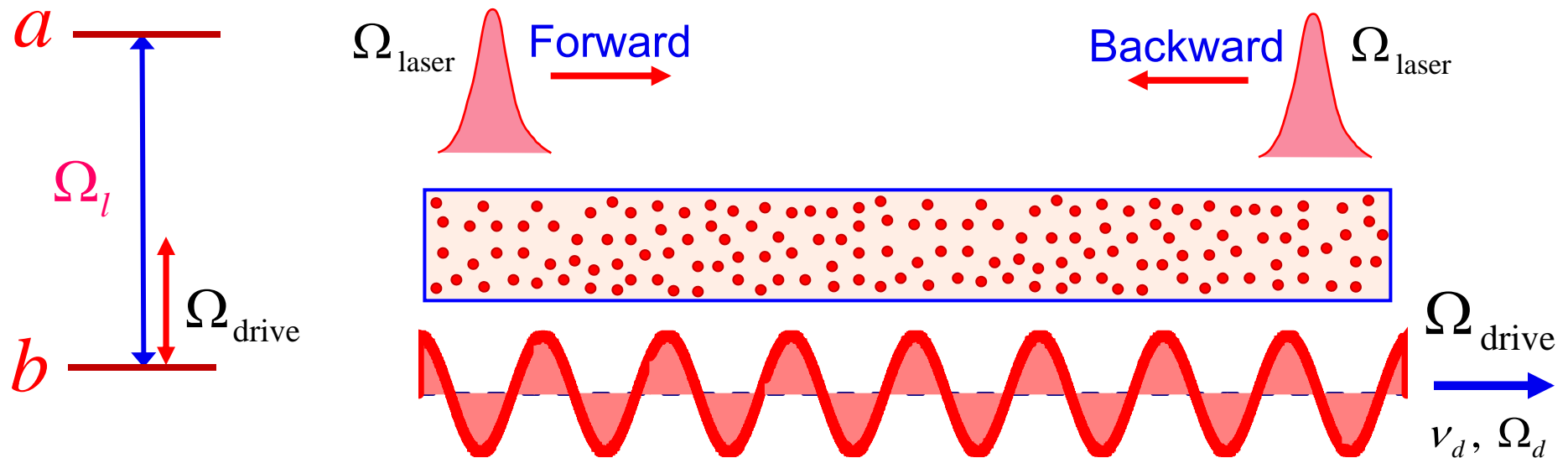
For 1<sup>st</sup> order resonance:

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

**G** – gain per unit time



# Propagating driving field: general analysis



Forward direction:

$$G=0$$

Backward direction:

If Stark shift is suppressed then

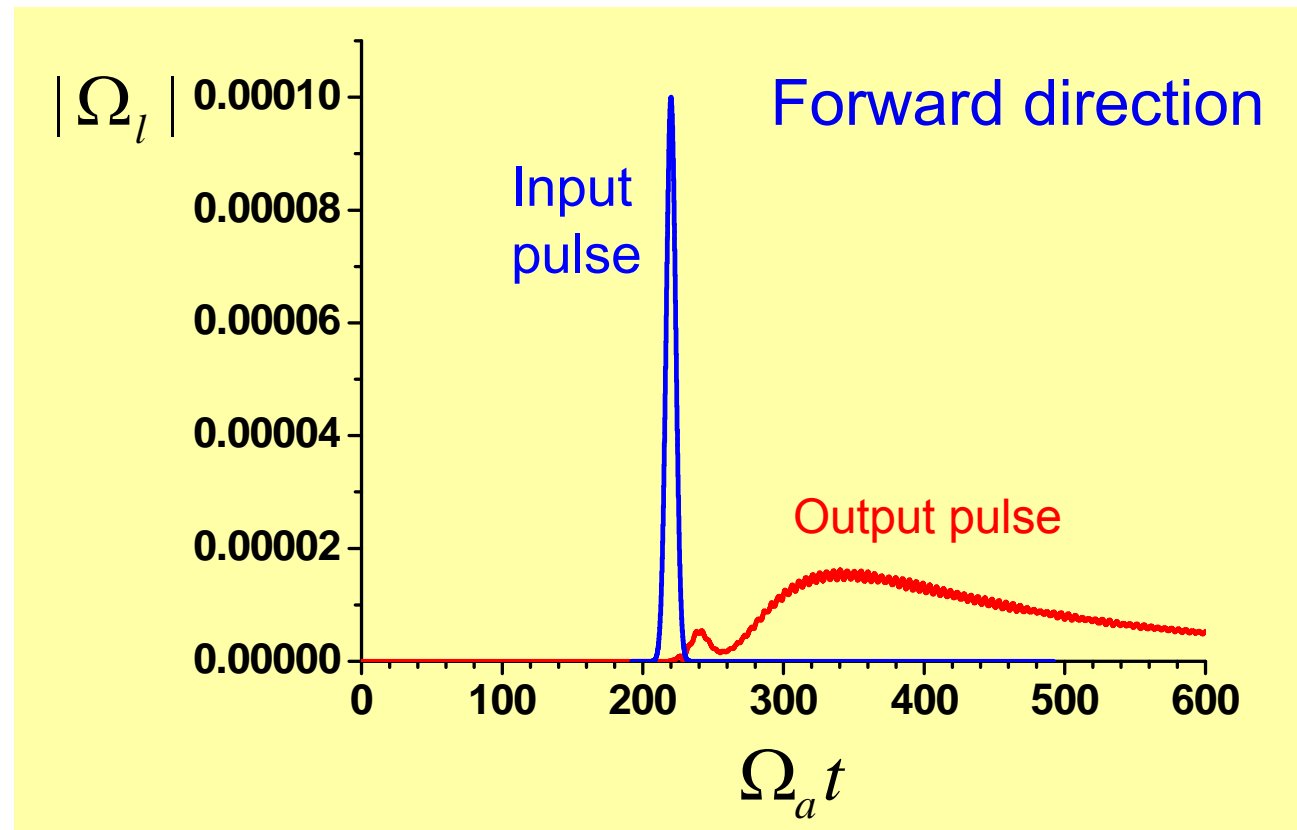
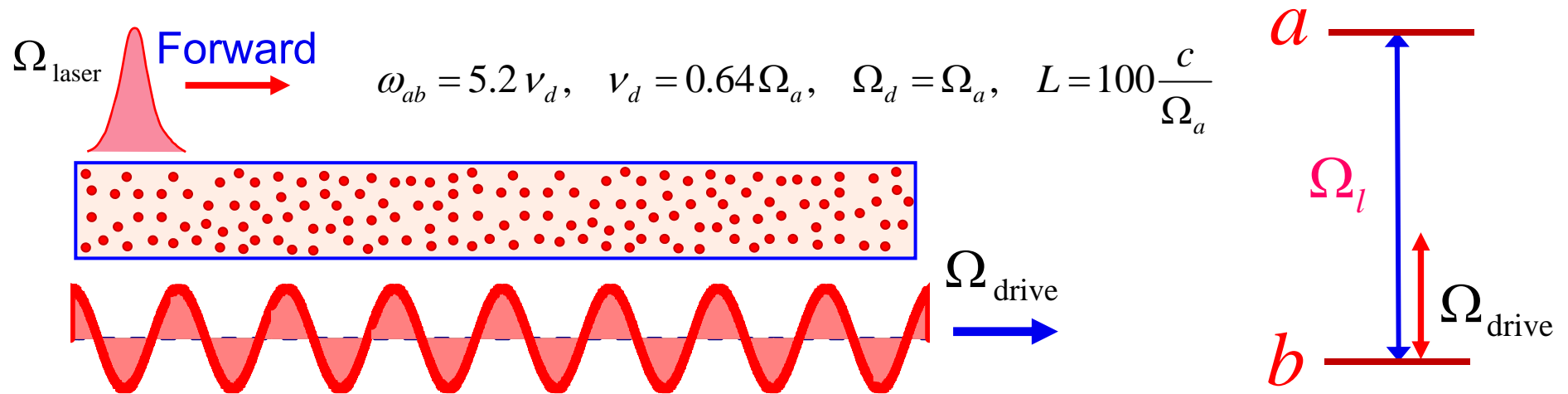
there is gain for

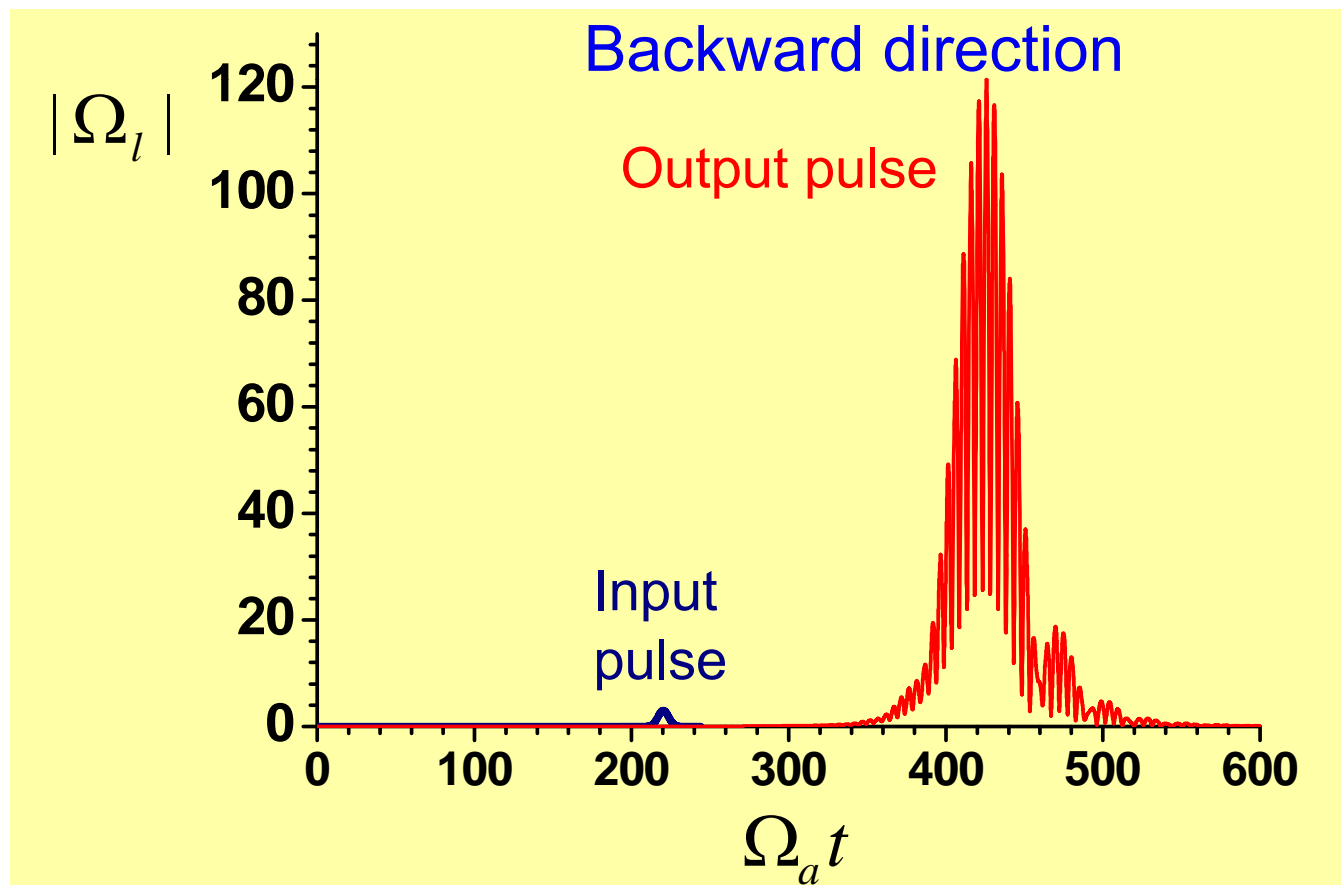
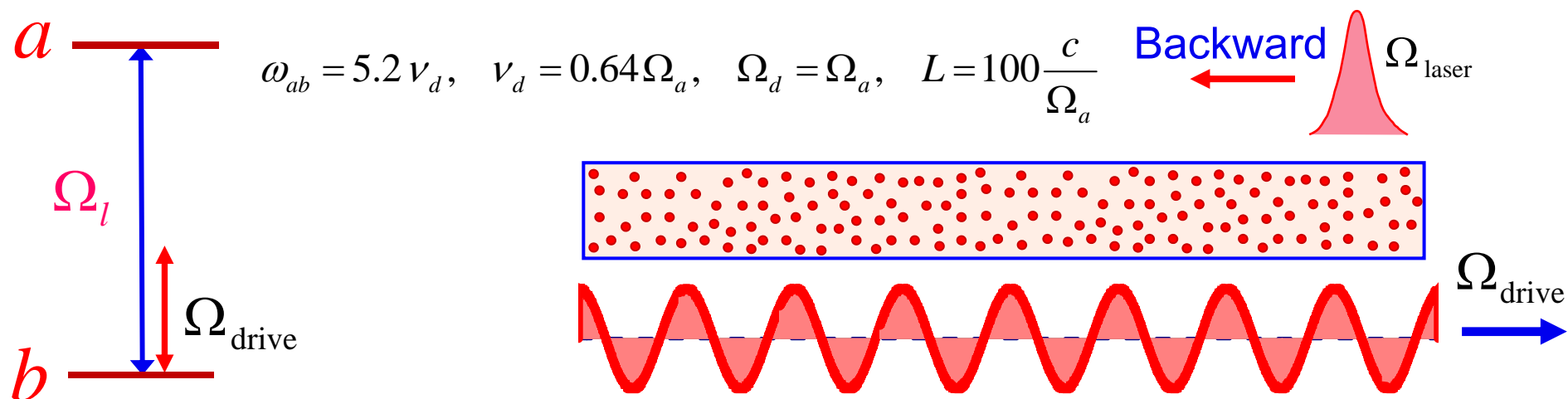
$$\nu_d > \frac{\Omega_a}{\sqrt{2}}$$

Gain per unit time:

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

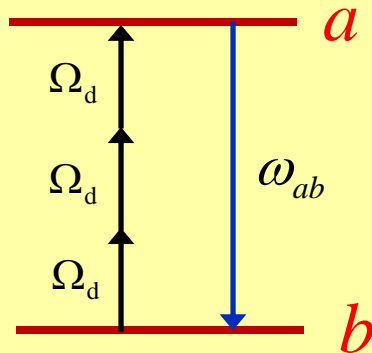
# Numerical solution of Maxwell-Schrödinger equations





# Comparison of atomic excitation mechanisms

## Multiphoton resonant excitation



Single atom phenomenon

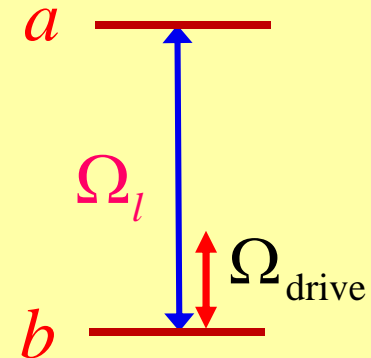
Multiphoton resonance with atomic transition frequency

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

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

High frequency light is emitted in the direction of driving field

## Collective parametric resonance



Collective effect

Resonance with collective frequency

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

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

Lasing occurs in the backward direction

# THE QASER REVISITED:

## Insights gleaned from analytical solutions to simple models\*

Marlan O. Scully

*Princeton University, Princeton, NJ 08544,*

*Texas A&M University,*

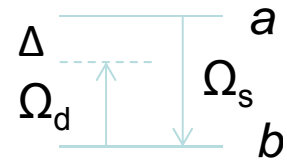
*College Station, TX 77843,*

*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 multi-photon Hamiltonian obtained by, e.g., a canonical transformation.

June 20, 2014

# SIMPLE GAIN EQUATION



$$\left. \begin{aligned} \dot{\Omega}_s &= -i\Omega_a^2 \rho_{ab} \\ \dot{\rho}_{ab} &= -i\Omega_s (1 - 2\rho_{aa}) \end{aligned} \right\} \begin{array}{l} \text{Photon Osc.} \rightarrow \ddot{\Omega}_s = (-i)^2 \Omega_a^2 (1 - 2\rho_{aa}) \Omega_s \\ \text{Atomic Osc.} \rightarrow \ddot{\rho}_{ab} = (-i)^2 \Omega_a^2 (1 - 2\rho_{aa}) \rho_{ab} \end{array}$$

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

$$X = \Omega_s \text{ or } \rho_{ab} \quad \text{SAME EQUATION SAME GAIN} \quad \text{Gain} = \frac{\Omega_a}{4} \left( \frac{\Omega_d}{\Delta} \right)^2$$



# Parametric harmonic oscillator (Mathieu equation)

$$\frac{\partial^2 x}{\partial t^2} + \Omega_a^2 [1 - \delta \cos(\nu_d t)] x = 0 \quad \delta \ll 1$$

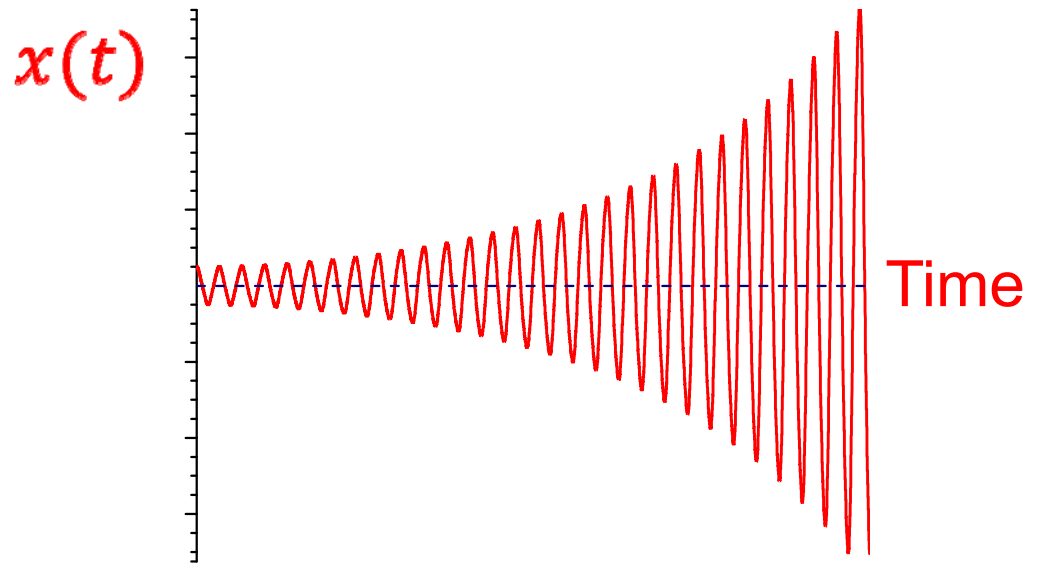
## Parametric resonance

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

For 1<sup>st</sup> order resonance:

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

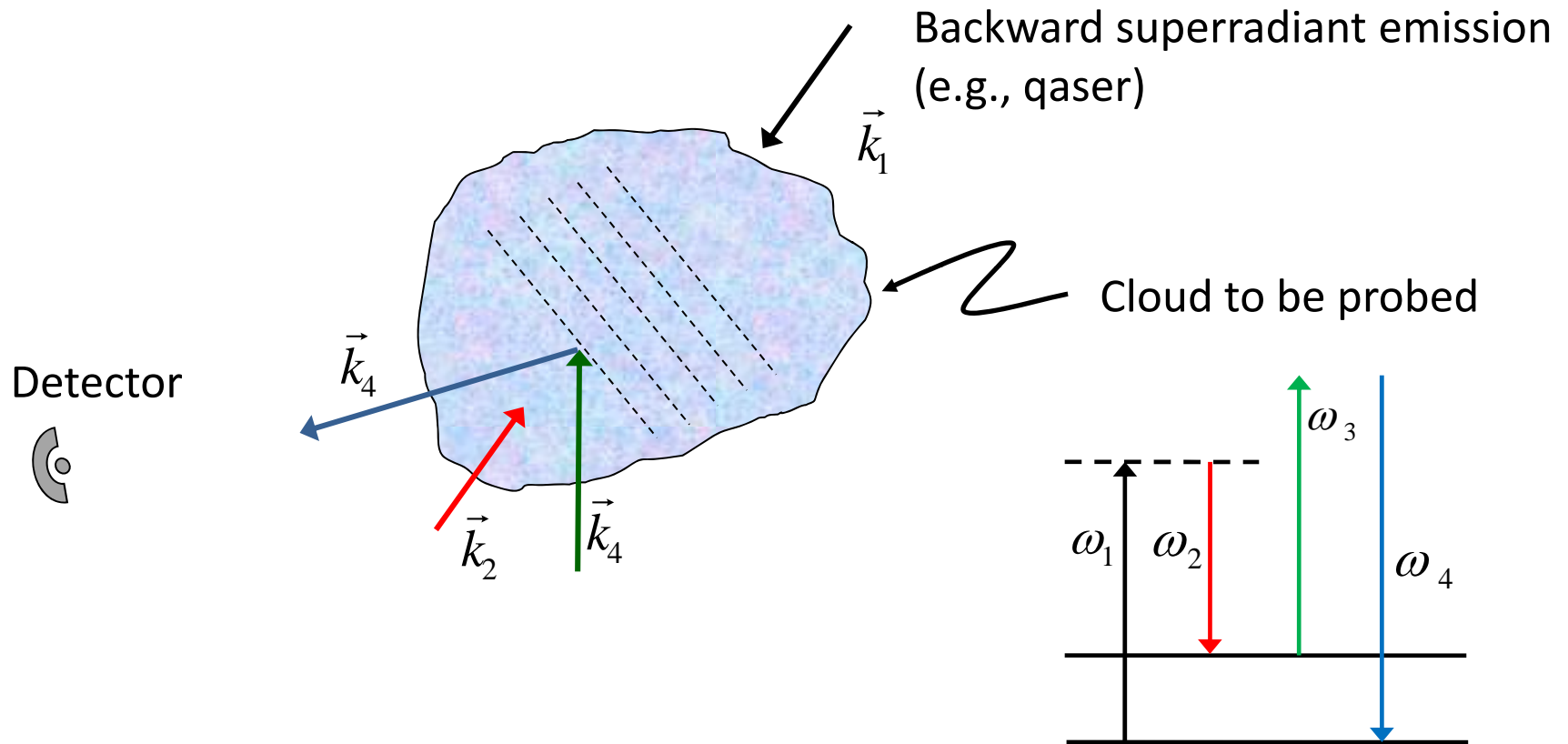
$G$  – gain per unit time



# **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

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

## 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)

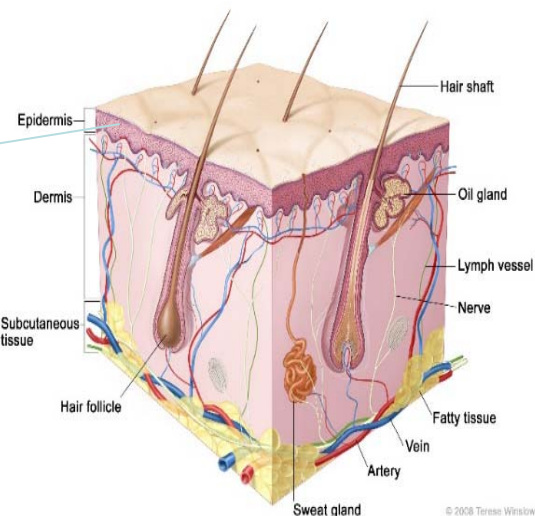
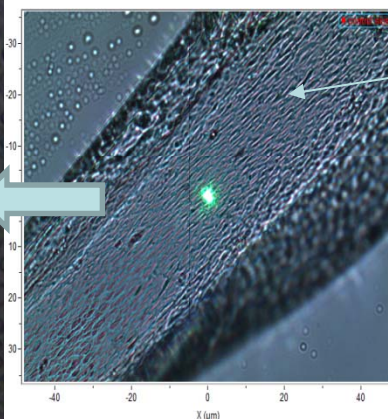
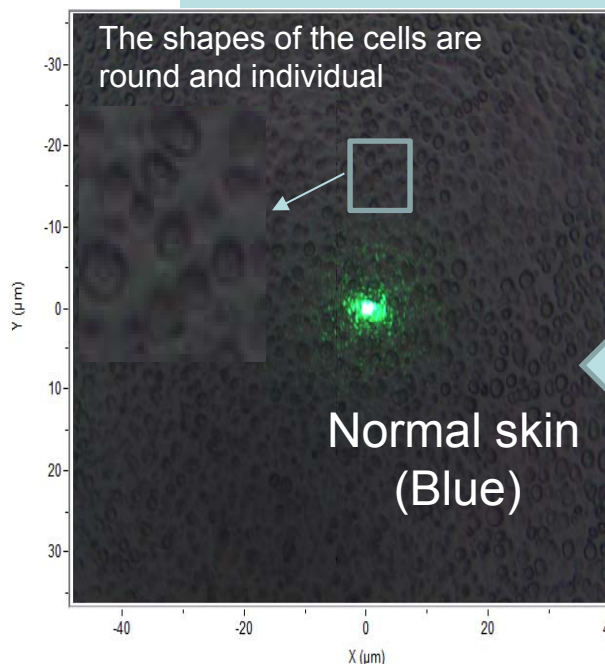
### 6) Summary

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

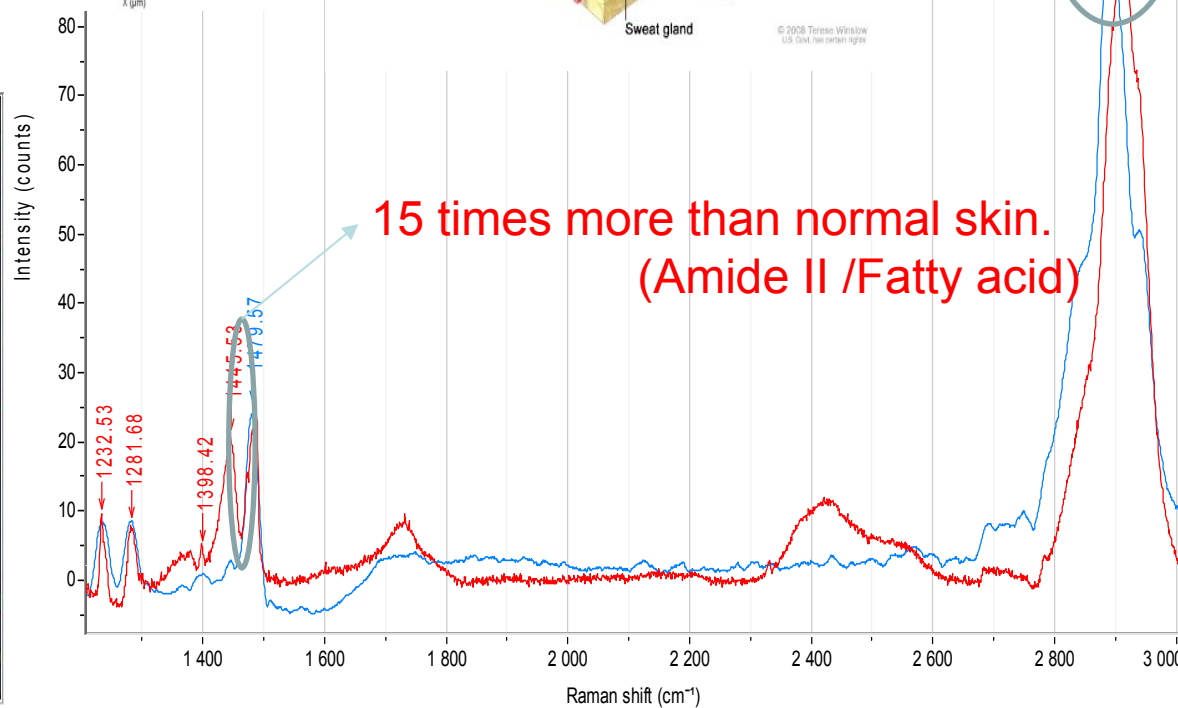
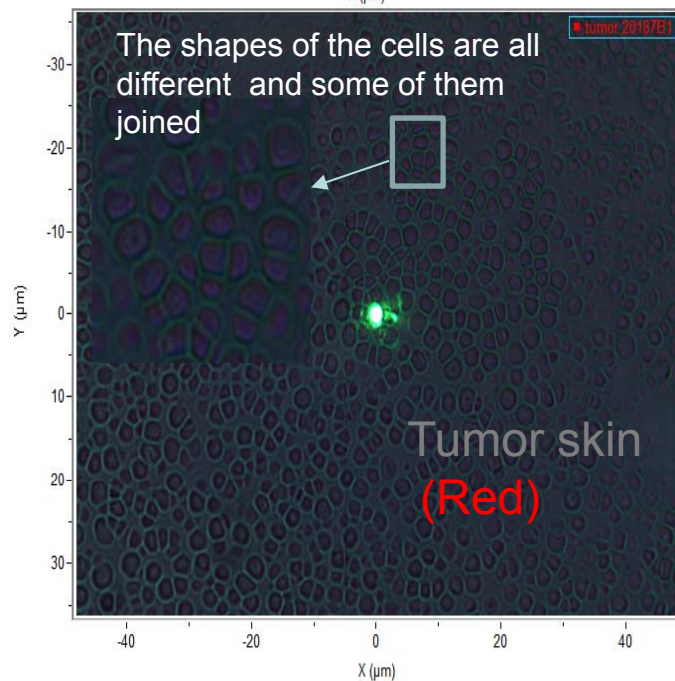
# A Raman Spectroscopic Study of skin tumors of Sinclair Piglets



# Main results on normal skin vs. tumor skin slides



Shifted 20 cm<sup>-1</sup>

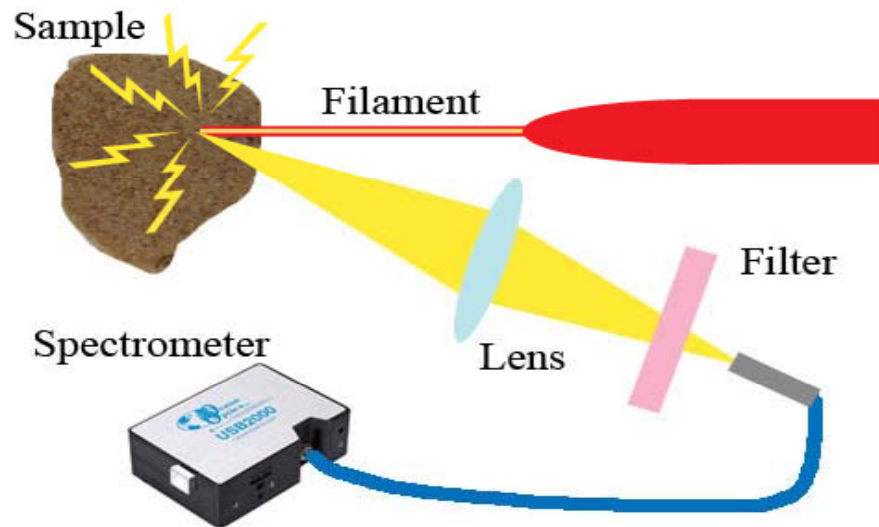


10/22/14, Nara



# Filament-induced breakdown spectroscopy (FIBS)

- Applications in precision agriculture, military, all areas of remote sensing
- Remotely detect and identify biological and mineralogical samples

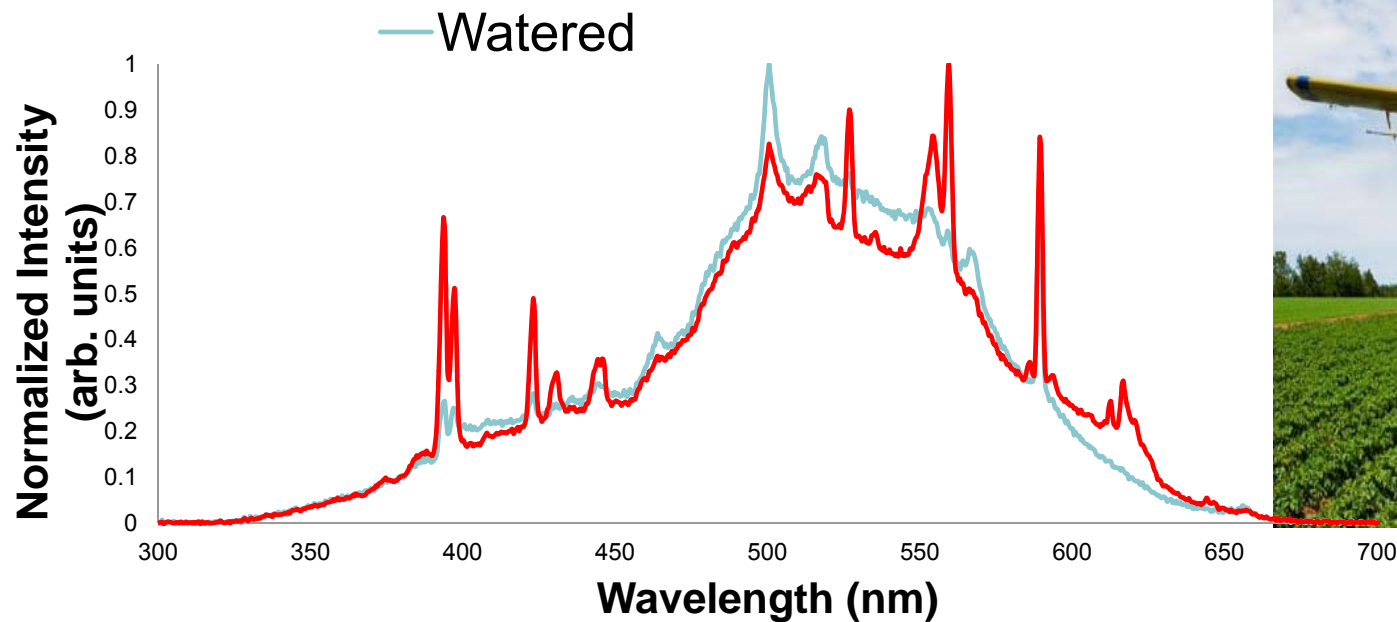


Aleksei  
Zheltkiov,  
Texas A&M,  
Moscow State

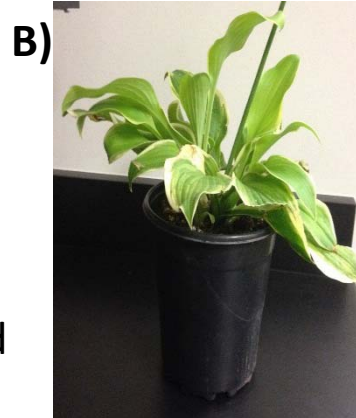


Courtesy of Tim Duckworth, US Navy

# Laser-Induced Breakdown Spectroscopy (LIBS) of Hosta Leaf



A) Hosta plant that is watered regularly.



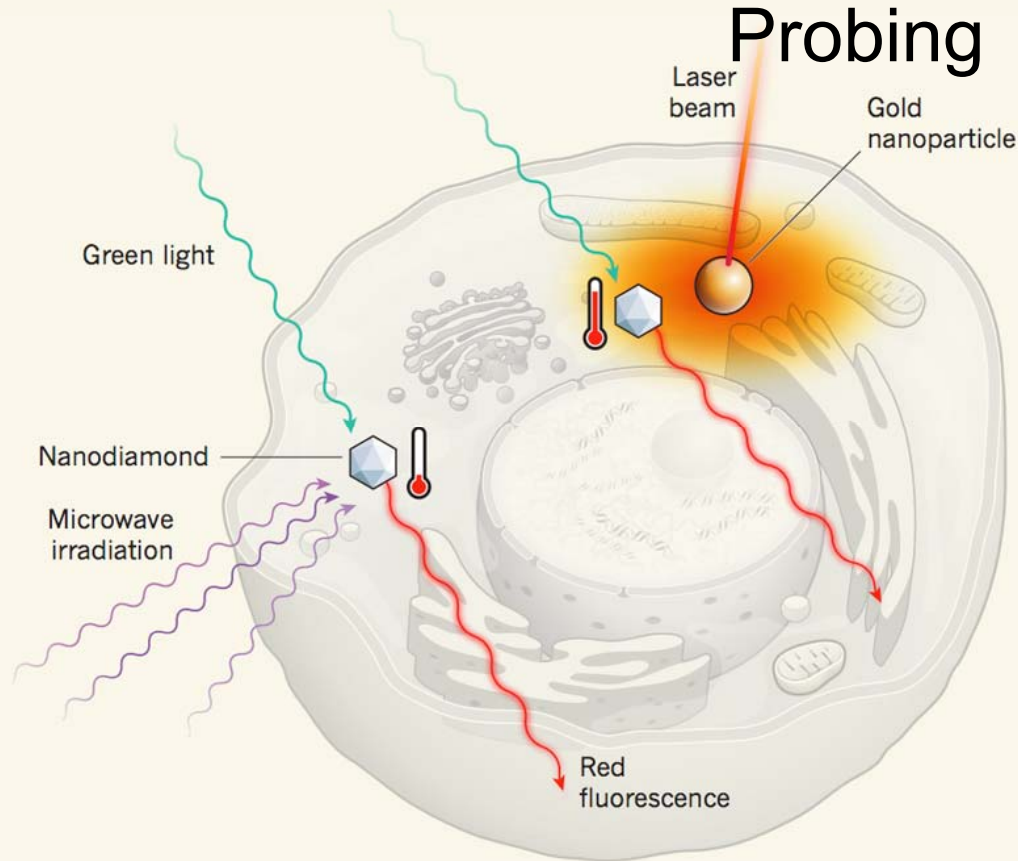
B) Hosta plant that has not been watered for 1 week.

Dmitri Voronine  
Texas A&M





# Quantum Nanosystems and Quantum Optical Probing



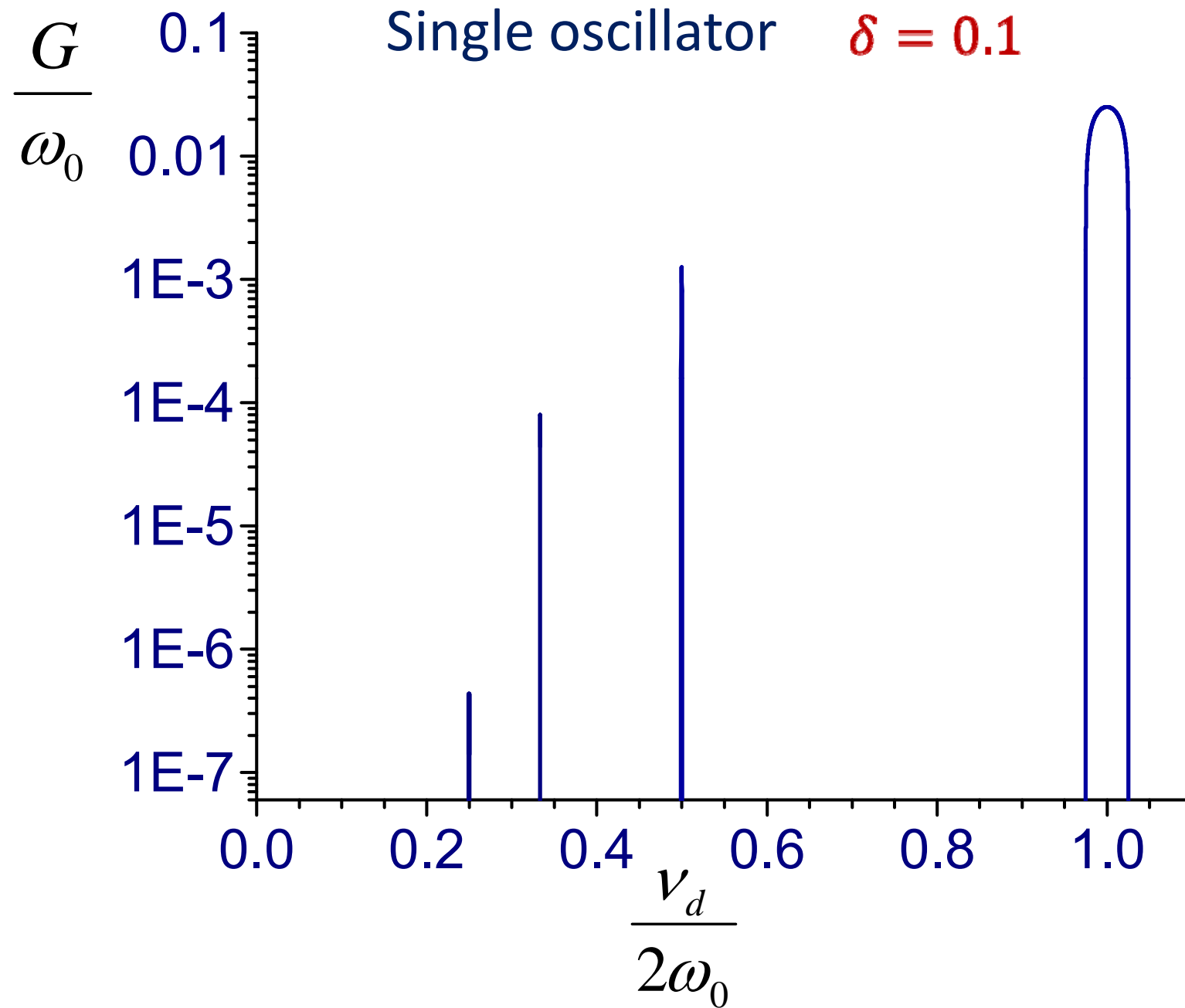
Mikhail  
Lukin,  
Harvard

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 $\omega_1, \omega_2, \omega_3, \dots$

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

$$\nu_d = \omega_2 - \omega_1 \quad \leftarrow \text{Difference combination resonance}$$

Example: Coupled parametric oscillators

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

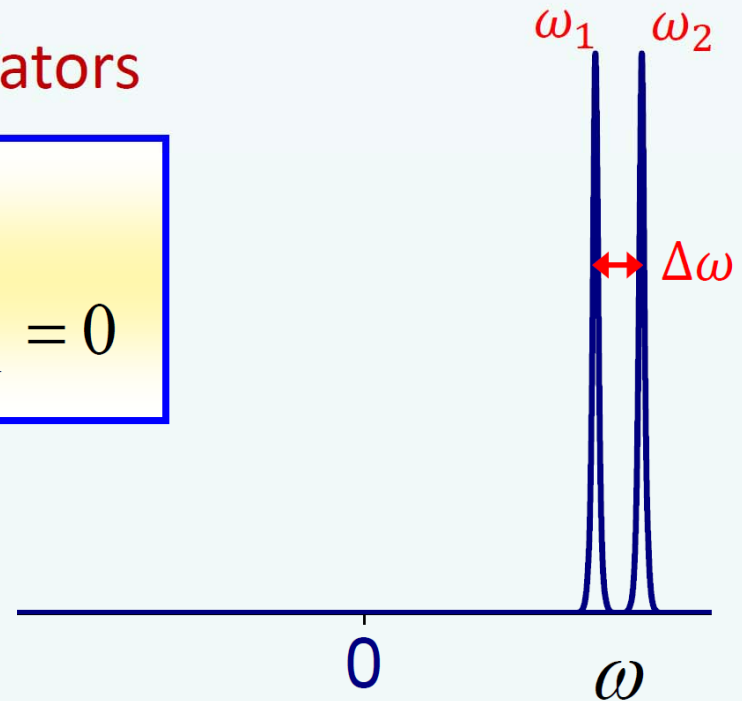
$$\ddot{x}_2 + \omega_0^2 x_2 - \Omega^2 [1 + \delta \cos(\nu_d t)] x_1 = 0$$

For 1<sup>st</sup> order resonance:

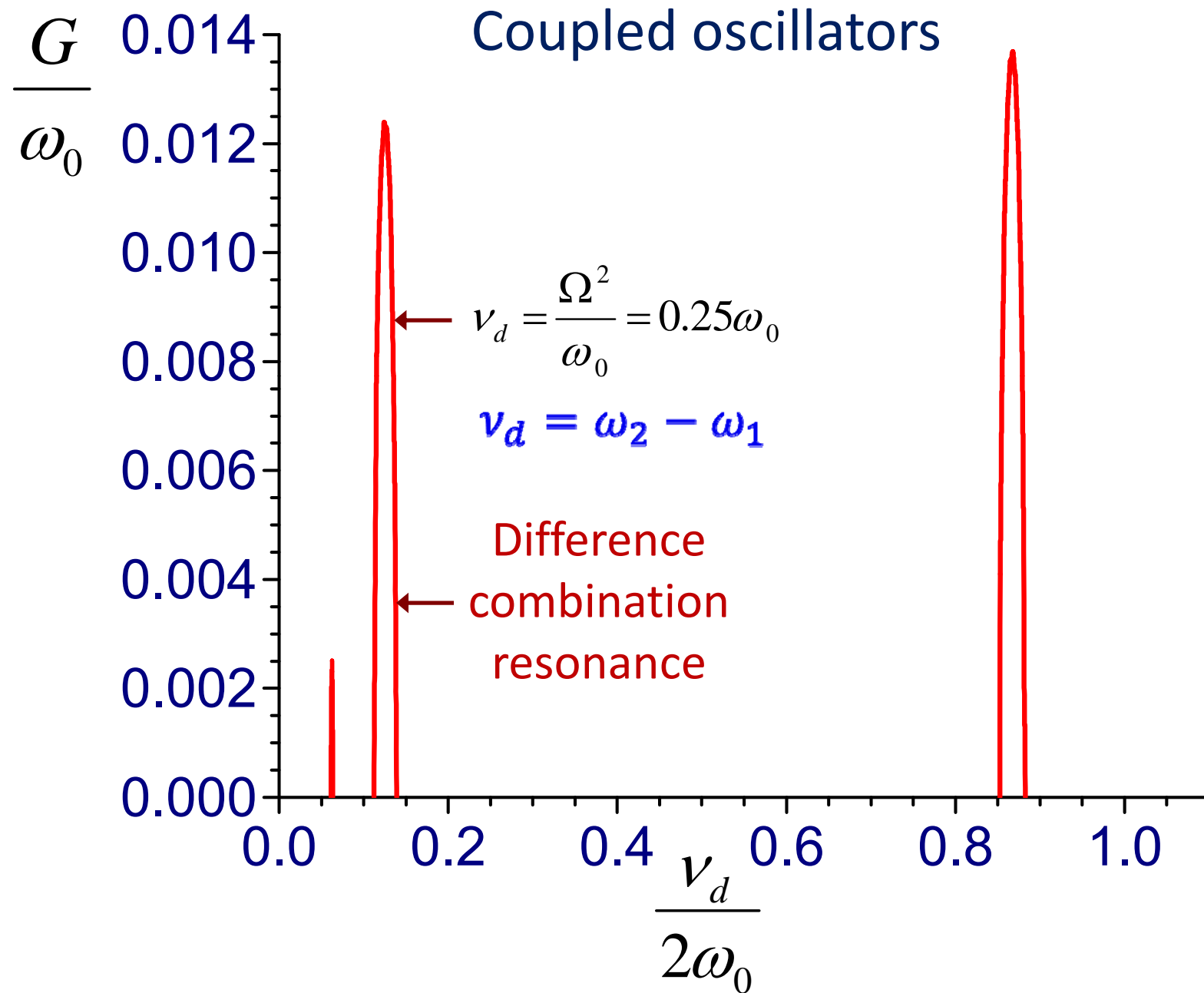
$$G \propto \delta \cdot \Delta\omega$$

We solve equations numerically with

$$\frac{\Omega^2}{\omega_0^2} = 0.25, \quad \delta = 0.4$$

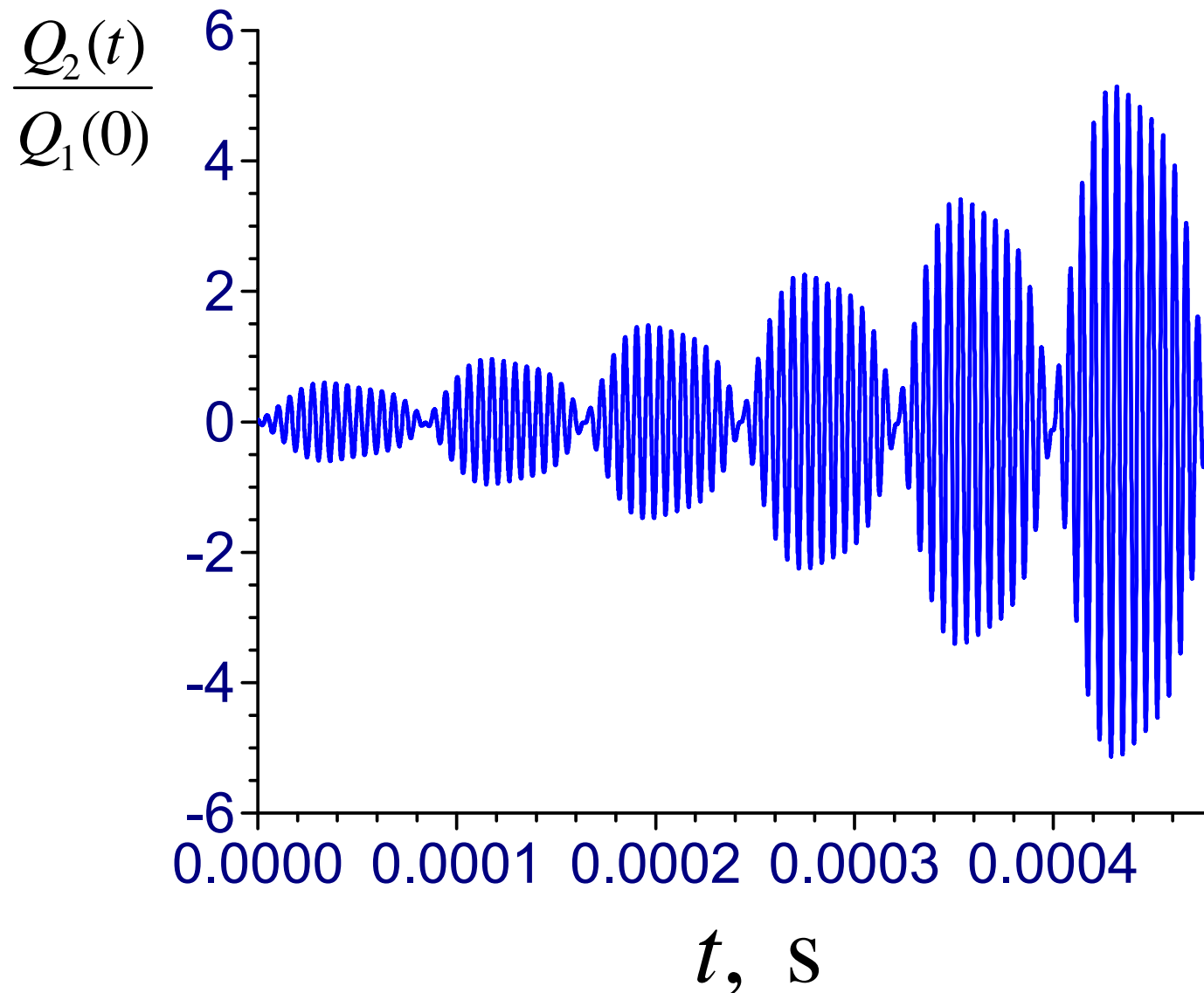


## Gain per unit time as a function of modulation frequency

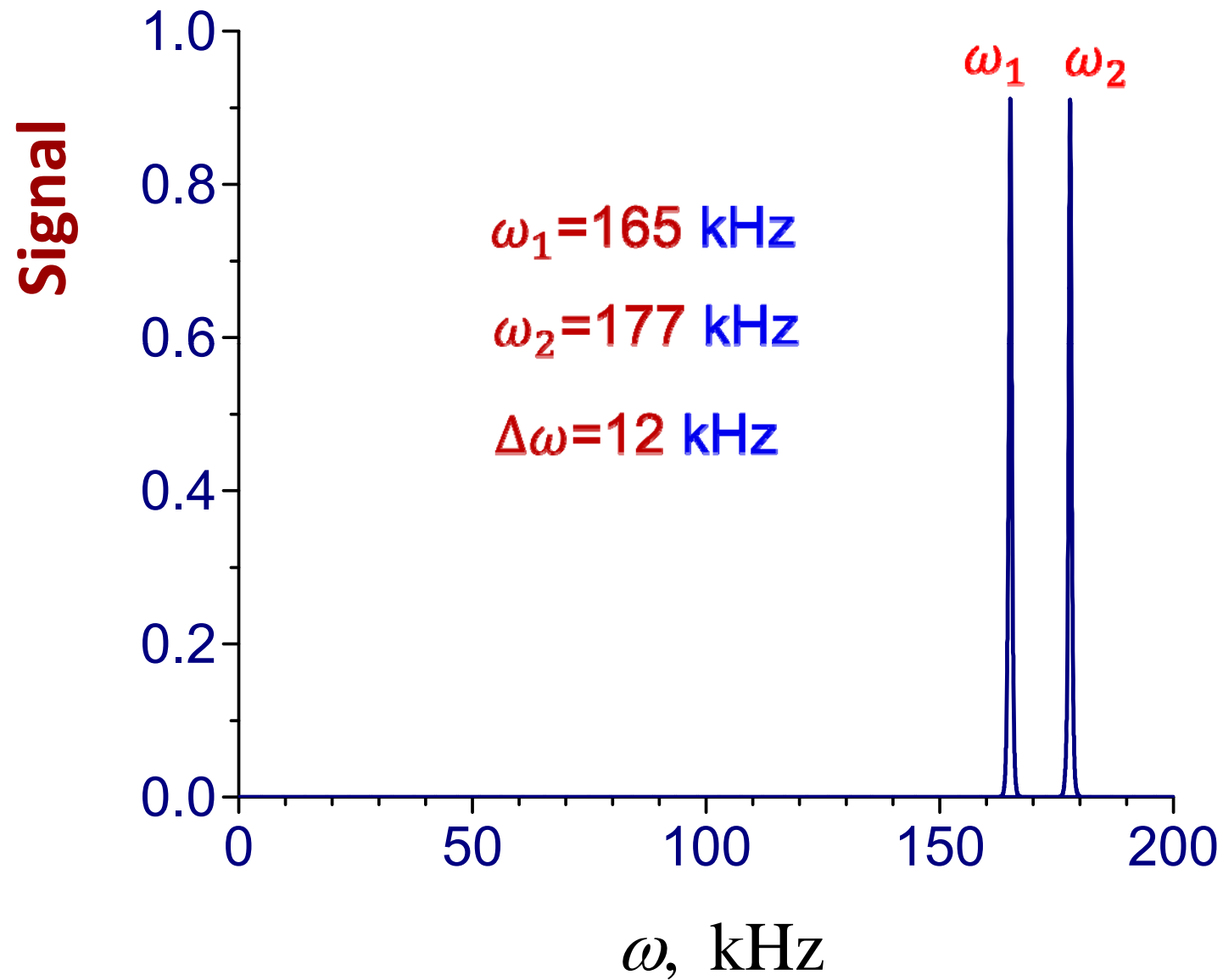


## Oscillation of charge on capacitor $C_2$ (Theory)

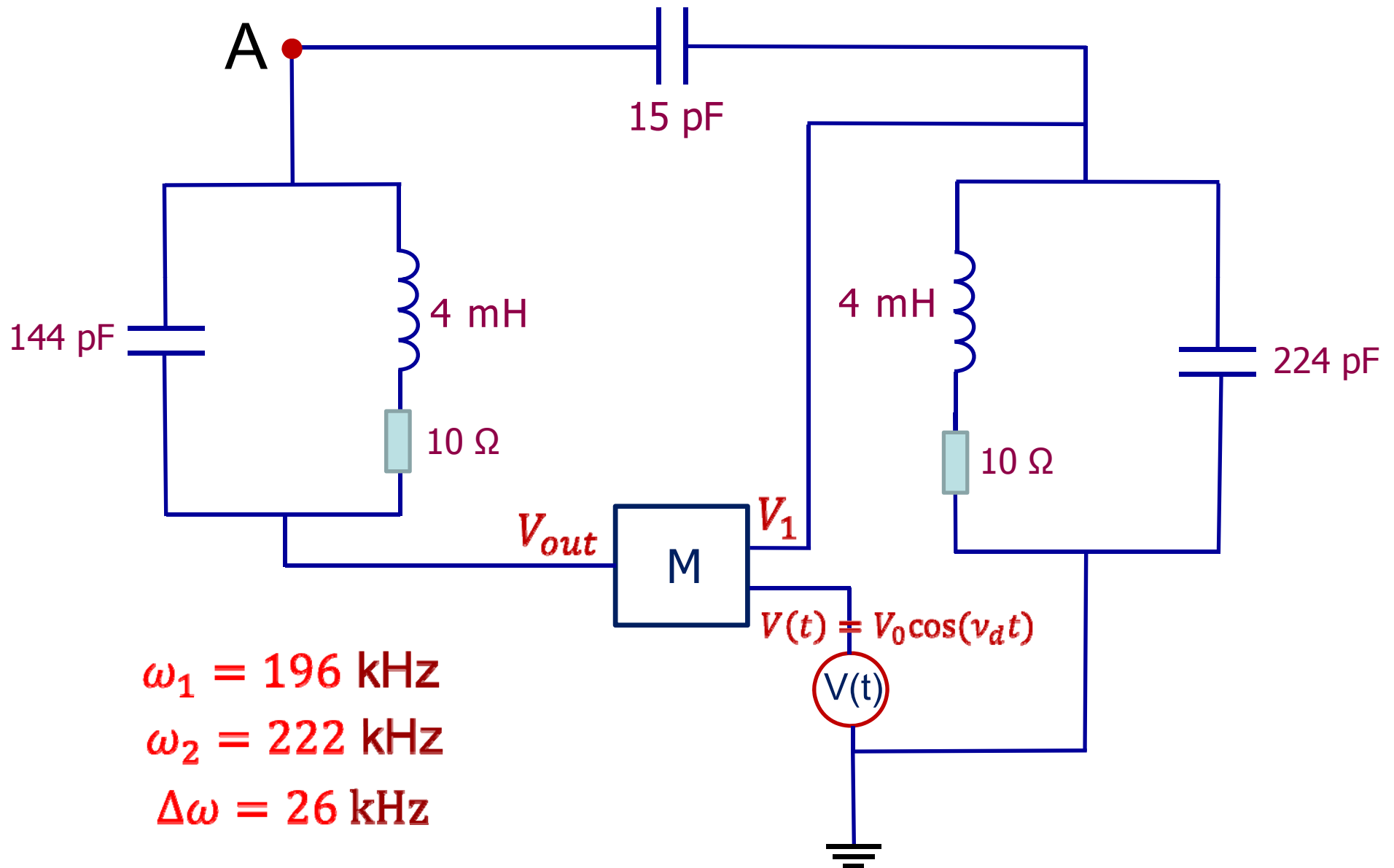
With drive  $\alpha V_0 = 0.1, \nu_d = 12 \text{ kHz}$

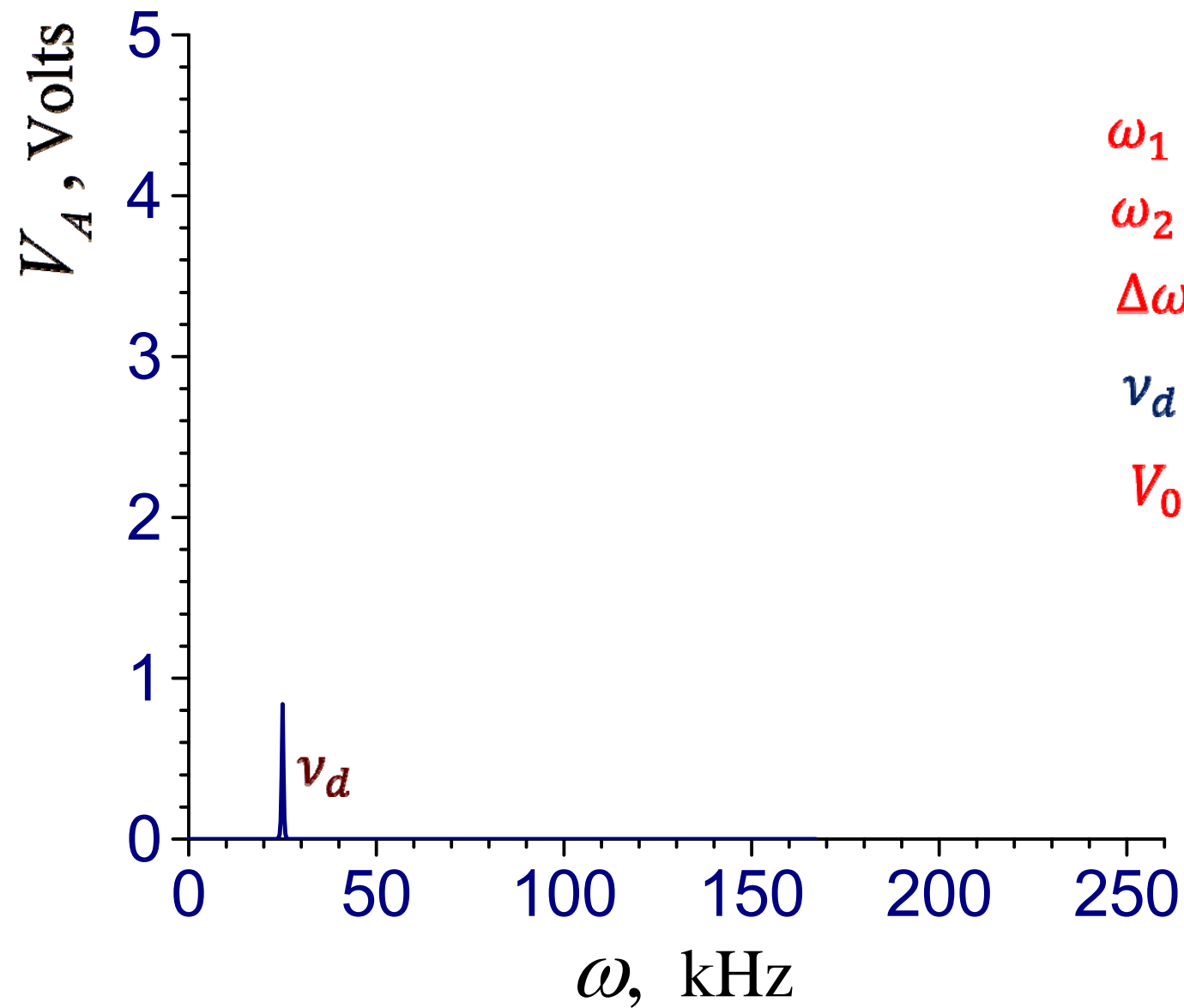


## In frequency domain (Theory)



# Experimental demonstration of difference combination resonance





$$\omega_1 = 196 \text{ kHz}$$

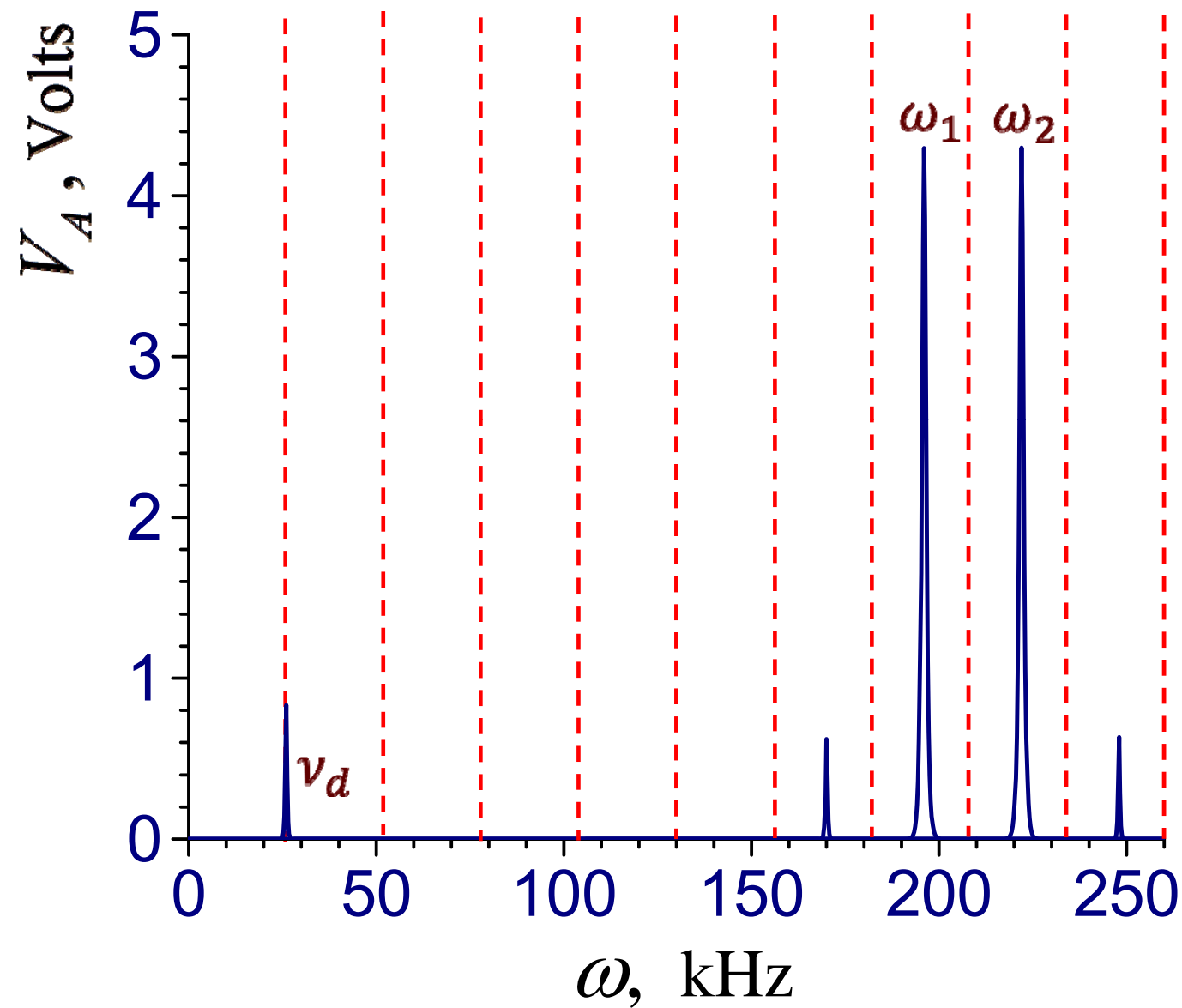
$$\omega_2 = 222 \text{ kHz}$$

$$\Delta\omega = 26 \text{ kHz}$$

$$\nu_d = 25 \text{ kHz}$$

$$V_0 = 0.9 \text{ V}$$





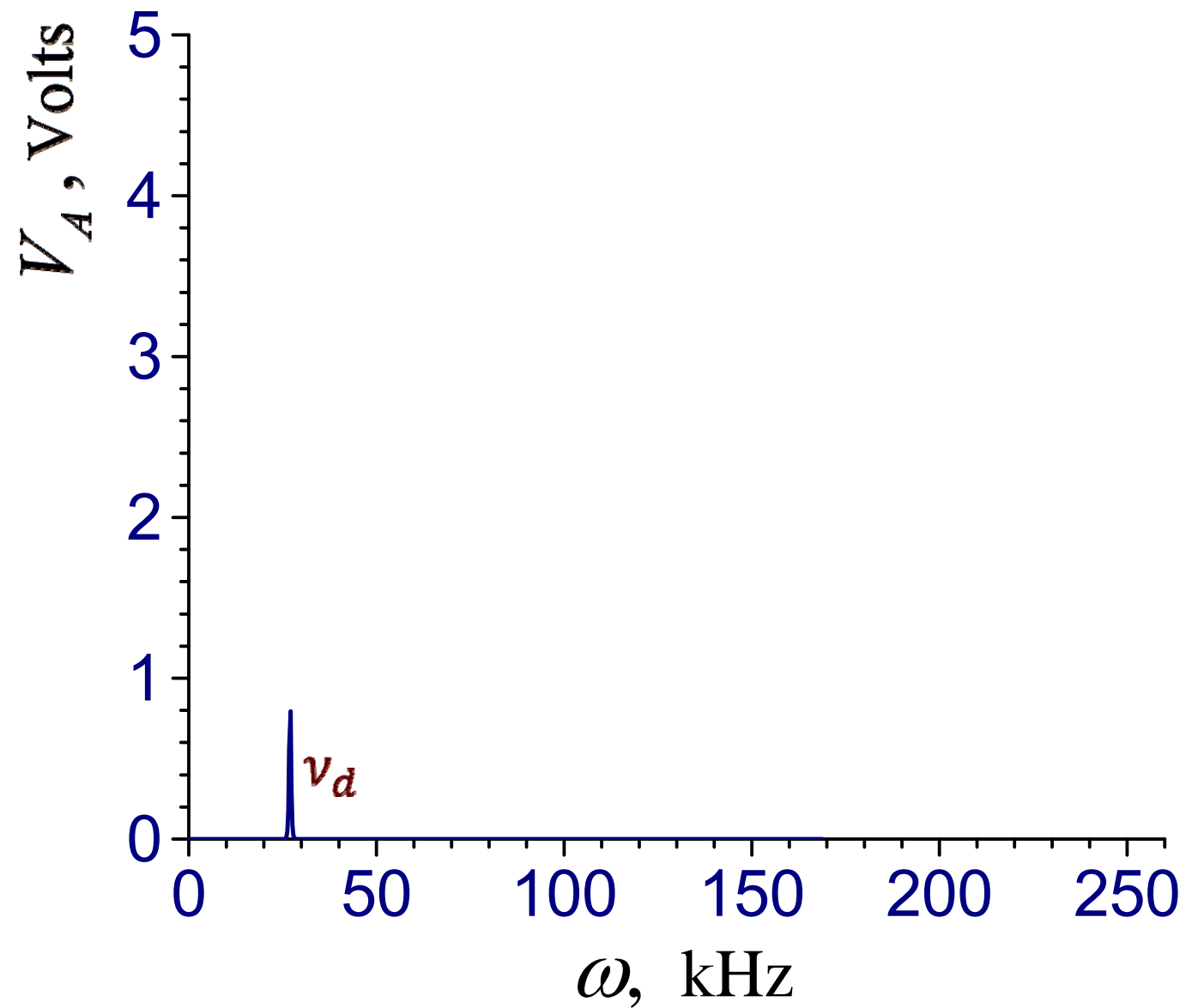
$$\omega_1 = 196 \text{ kHz}$$

$$\omega_2 = 222 \text{ kHz}$$

$$\Delta\omega = 26 \text{ kHz}$$

$$\nu_d = 26 \text{ kHz}$$

$$V_0 = 0.9 \text{ V}$$



$$\omega_1 = 196 \text{ kHz}$$

$$\omega_2 = 222 \text{ kHz}$$

$$\Delta\omega = 26 \text{ kHz}$$

$$\nu_d = 27 \text{ kHz}$$

$$V_0 = 0.9 \text{ V}$$

