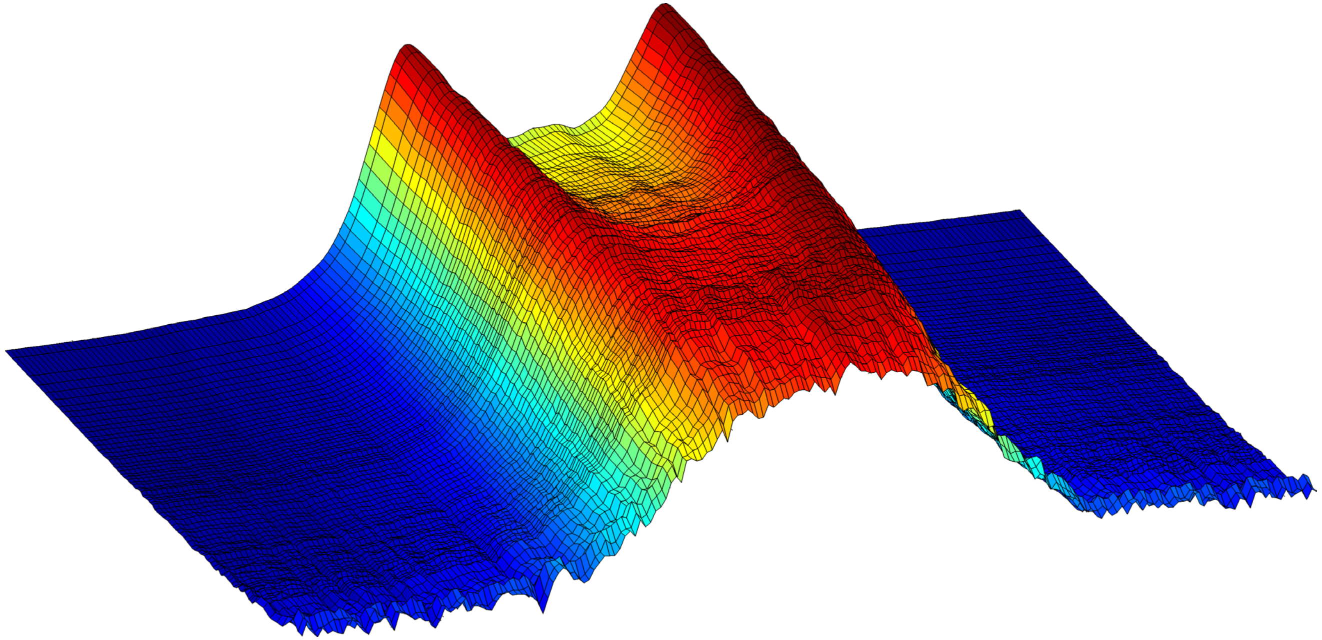

Low (and high) temperature plasma Laser Diagnostics

Alexandros Gerakis

Princeton Plasma Physics Laboratory
Introduction to Fusion Energy and Plasma Physics Course
June 15th, 2023

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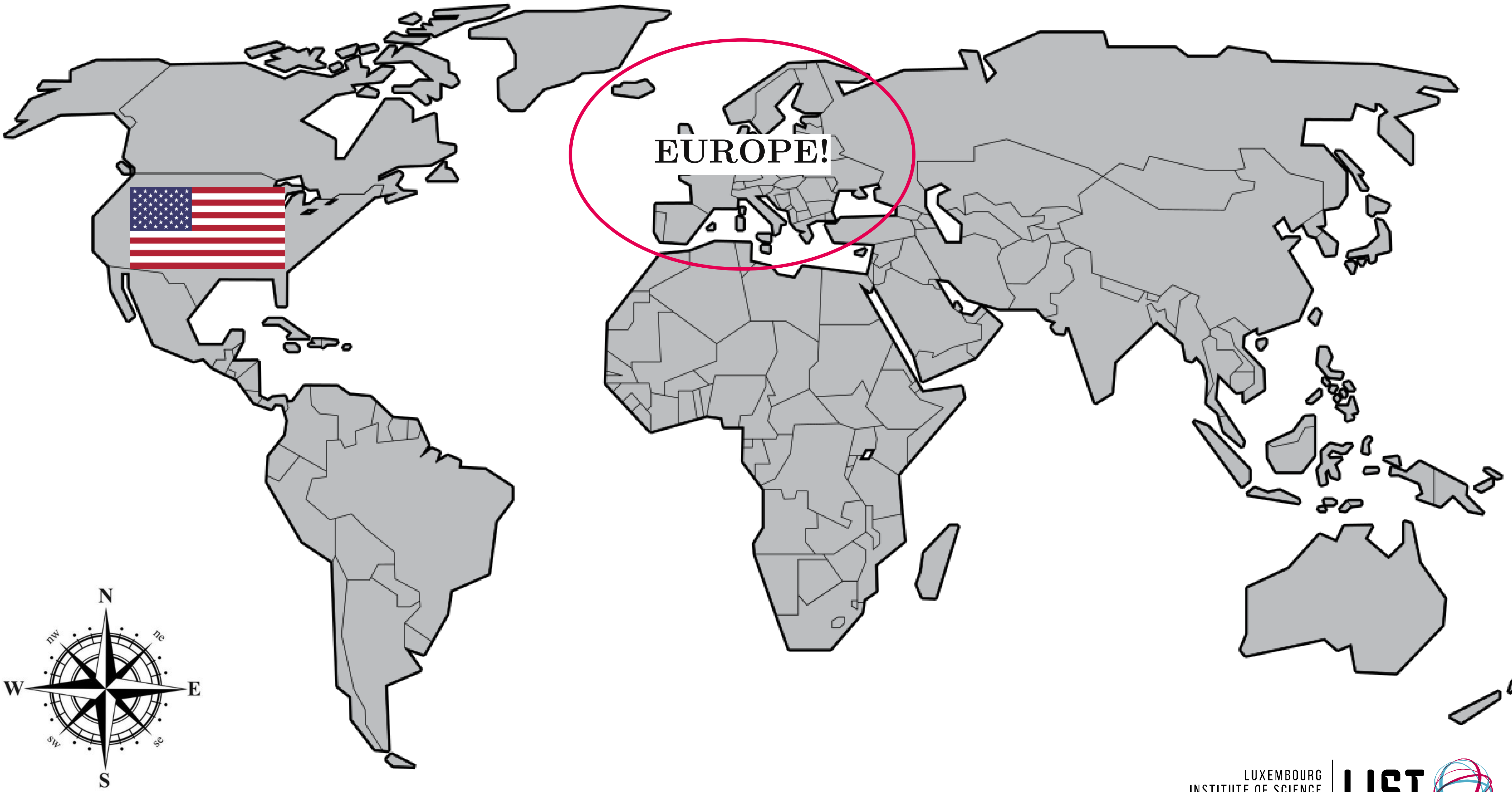

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AT THE HEART OF BUSINESS

General Questions about Luxembourg

- It is not in Germany
- Yes, it is its own country.
- No, it is not a kingdom but a Grand Duchy (the last one in the world).
- One of the founding members of the EU.
- It has three(!) official languages:
  - ❑ English is not one of them
  - ✓ Luxembourgish (yes, it is a thing)
  - ✓ German
  - ✓ French
- Population is roughly 615.000
- Almost same size as Rhode Island
- 260 times smaller than Texas.

LUXEMBOURG
LET'S MAKE IT HAPPEN

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THE NEW RESEARCH AND TECHNOLOGY ORGANIZATION

Created in 2015
Active in 4 fields

- ENVIRONMENT
- INFORMATICS
- MATERIALS
- SPACE

LIST

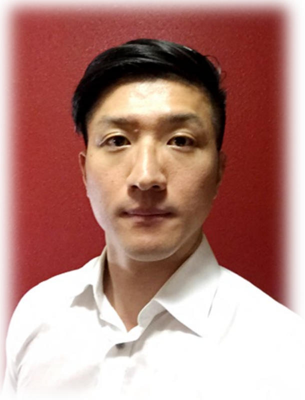
WHO I AM


PPPL
Luxembourg National Research Fund
ABOUT US
FUNDING
INNOVATION & INDUSTRY PARTNERSHIPS
INTERNATIONAL COOPERATION
SCIENCE IN SOCIETY
Home | Our News | Dr Alexandros Gerakis awarded FNR ATTRACT Fellowship
Dr Alexandros Gerakis awarded FNR ATTRACT Fellowship
18 Feb 2022
The FNR has awarded a FNR ATTRACT Fellowship to Dr Alexandros Gerakis for an innovative laser technology project at the Institute of Science and Technology (LIST). Dr Gerakis' AT doted with 2 MEUR over five years.
HERIOT WATT UNIVERSITY
University of St Andrews
LIST
Single-shot coherent
Diving deep into the theory of plasma physics
August 8, 2019 | By Jan McHarg
Aerospace Engineering
National Science Foundation (NSF)
Research
Drs. Alexandros Gerakis and Kentaro Hara | Image: Igor Kraguljac
With a $300,000 grant from the National Science Foundation, a team of researchers at Texas A&M University is diving deep into the physics of plasma to question the fundamental theory of local thermodynamic equilibrium (LTE) in arc discharges.

ACKNOWLEDGEMENTS

Optical Probing
&
Manipulation Group

TEXAS A&M UNIVERSITY



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Dr. Kentaro Hara

Stanford University

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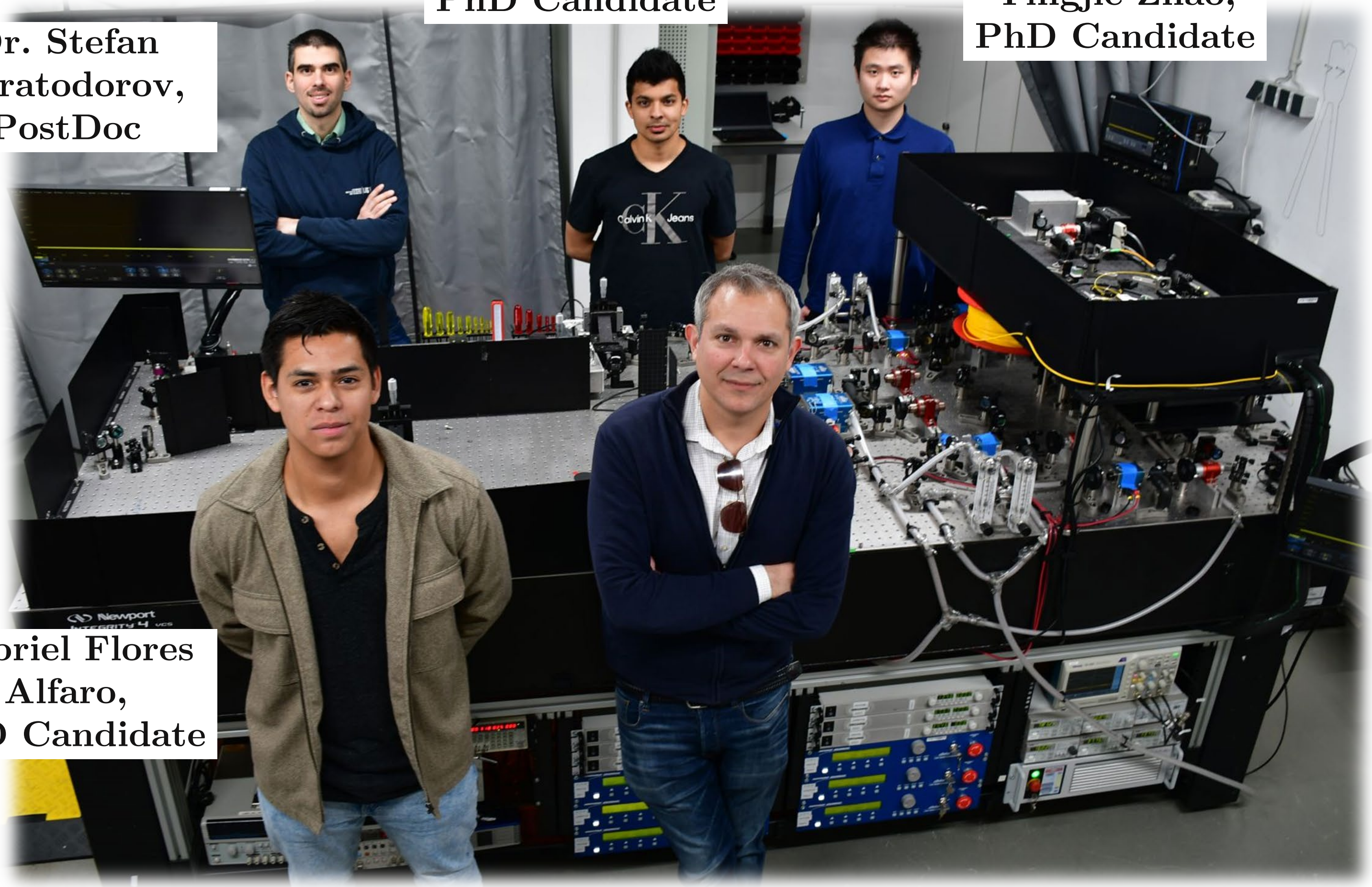
ACKNOWLEDGEMENTS



Luxembourg National
Research Fund

ATTRACT

ATTRACTING
OUTSTANDING YOUNG
RESEARCHERS FROM
ABROAD

CORE MULTI-ANNUAL
THEMATIC RESEARCH PROGRAMME

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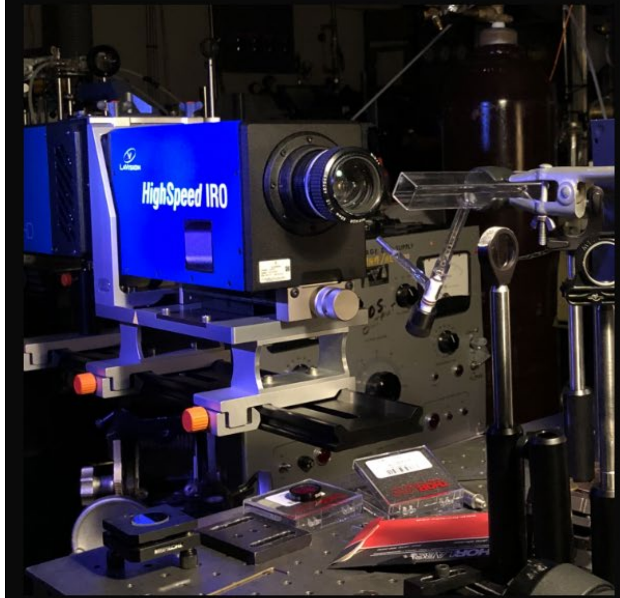
DISCLAIMER NO.1

This is <u>not</u> an exhaustive list

Research Capabilities



Advanced Diagnostics

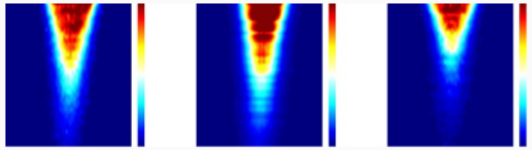
- Atomic and molecular species, positive and negative ions
  - Laser-Induced Fluorescence (LIF) and Two-Photon Absorption LIF (TALIF)
    - Nanosecond LIF/TALIF
    - Femtosecond TALIF
    - CW LIF for measurements of Ar and Xe ion and atom VDFs
  - Radar Resonant Enhanced Multi-Photon Ionization (Radar REMPI)
- Characterization of plasma, chemical composition and dynamic behavior
  - Optical Emission Spectroscopy (OES)
  - Fourier Transform Infrared Spectrometry
  - Fast Imaging
  - Electrostatic Quadrupole Mass Spectrometer and Energy Analyzer
  - Electrostatic probes
- Electron velocity distribution function, gas flow velocity and temperature
  - Thomson, Raman, and Rayleigh scattering
  - Hybrid Coherent Anti-Stokes Raman Scattering (Hybrid CARS)
  - Femtosecond Laser Electronic Excitation Tagging (FLEET)
- Electric field and space potential
  - Electric Field-Induced Second Harmonic Generation (E-FISH)
  - Electrostatic probes
- Nanoparticle Diagnostics
  - Laser-Induced Incandescence (LII)
  - Laser-Induced Breakdown Spectroscopy (LIBS)
  - Coherent Rayleigh-Brillouin Scattering (CRBS)
- Surface Diagnostics
  - Surface Second Harmonic Generation (SHG)
  - Electron-induced Secondary Electron Emission and Surface Charging Measurements
  - Surface Potential Measurements with the Kelvin Probe Diagnostic

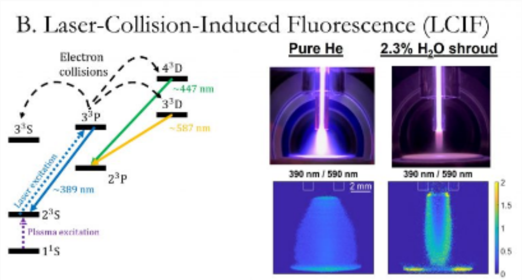
https://pcrf.princeton.edu/capabilities/#diagnostics

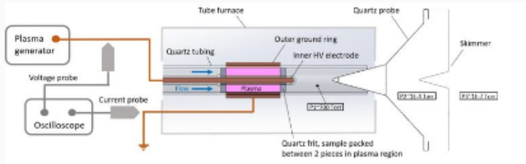
Capabilities

- Multiple femtosecond, picosecond, nanosecond and CW lasers spanning < 190 nm and exceeding 10 mm for interrogating dynamic plasma and reactive environments.
  - LCIF for electron densities and LIF-Dip for electric fields.
  - 2D-CARS and SFG for gas and surface phase interrogation
  - Multi-pulse (10 kHz to 100 kHz) laser cluster for high speed LIF and Rayleigh
- High speed imaging capabilities from <200 ps gated cameras, multi-frame framing cameras and high-speed CMOS cameras.
- VUV and FTIR spectrometers for spectroscopy.
- High resolution tandem and molecular beam mass spectroscopy for interrogating gas phase chemistries occurring in multi-atmosphere.

Broad range of plasma generating capabilities spanning vacuum to atmospheric pressures and beyond.







https://www.sandia.gov/prf/plasma-research-facility/capabilities/

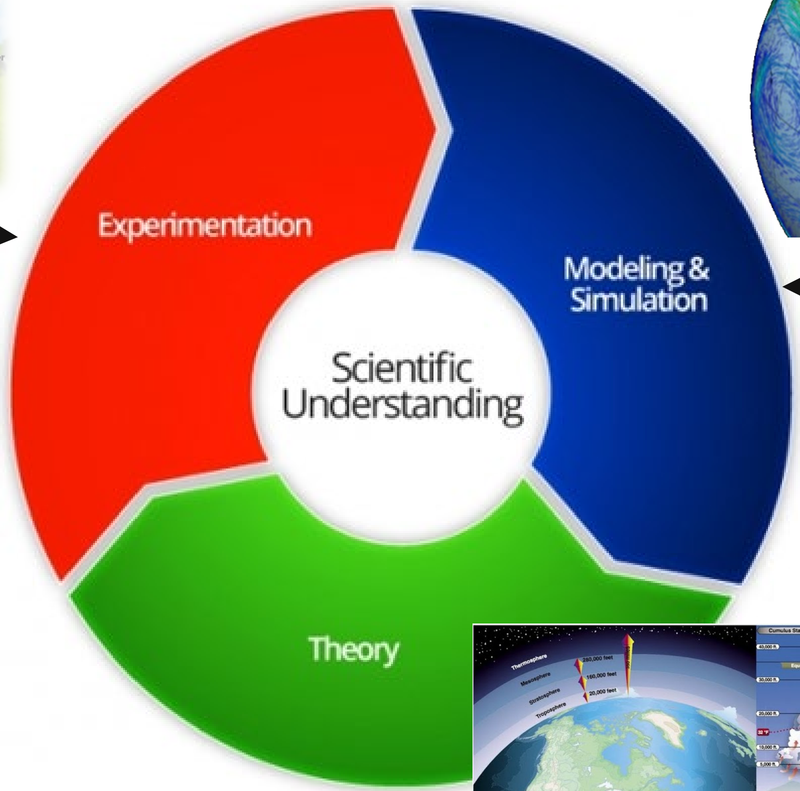
DISCLAIMER NO.2

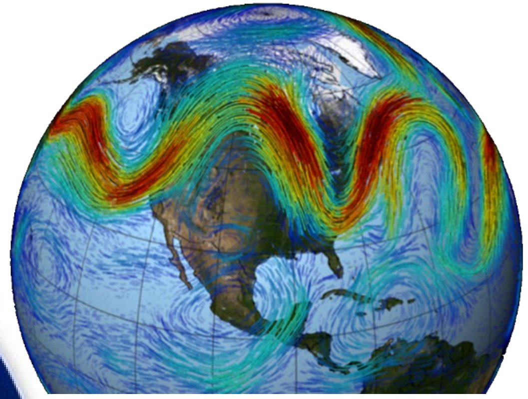
I will be going fast ☺

WHY CARE ABOUT METROLOGY?

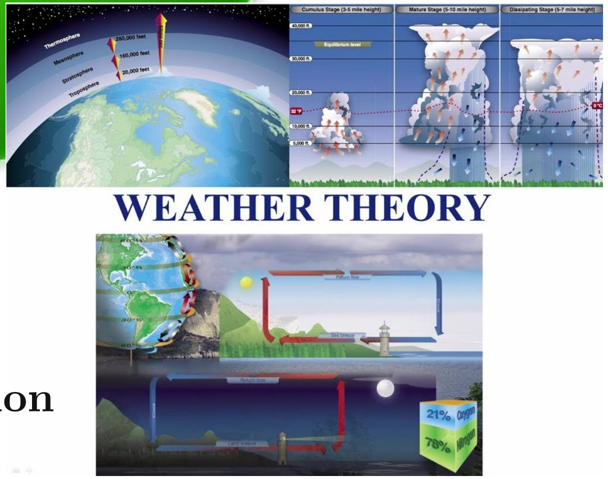
<u>Metrology:</u>
Tools used to measure/quantify a process

Metrology is defined by the International Bureau of Weights and Measures (BIPM) as "the science of measurement, embracing both experimental and theoretical determinations at any level of uncertainty in any field of science and technology".





Verification



Validation

From https://www.energy.gov/ne/articles/role-modeling-and-simulation-scientific-discovery

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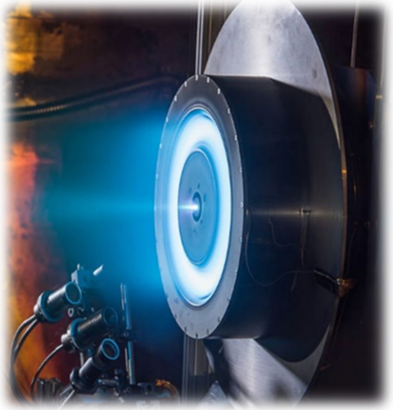
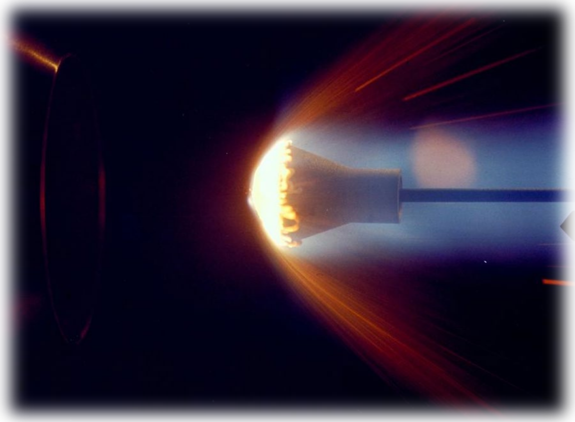
WHY CARE ABOUT LTP METROLOGY?



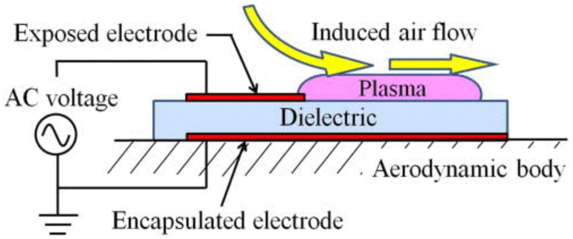
Hall Thruster

https://en.wikipedia.org/wiki/Hall-effect_thruster



Arc Jets

https://www.nasa.gov/centers/ames/multimedia/images/2008/A-28916-3.html

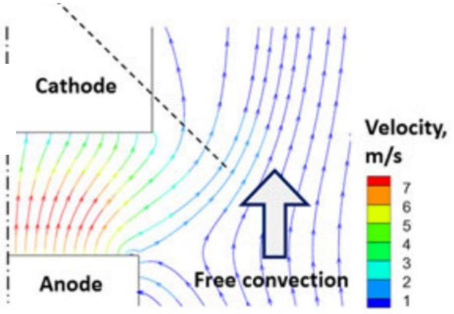


Dielectric Barrier Discharges

Nishida, Hiroyuki, Taku Nonomura, and Takashi Abe. "Three-dimensional simulations of discharge plasma evolution on a dielectric barrier discharge plasma actuator." *Journal of Applied Physics* 115, no. 13 (2014): 133301.

Magnetron sputtering

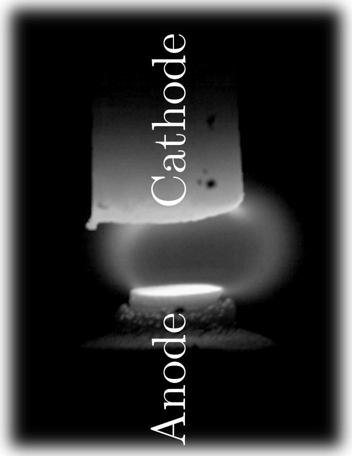
https://intlvac.com/News-Resources/ArticleID/5/What-is-Sputtering

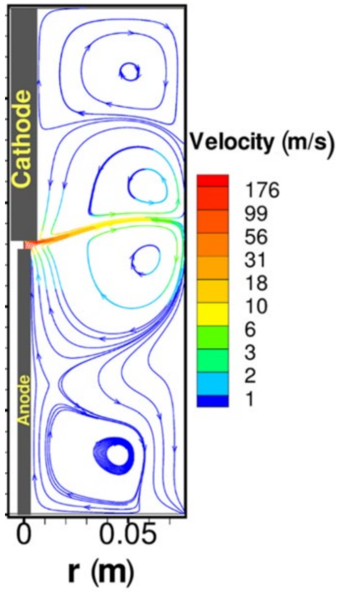


Flow pattern (velocity streamlines)
(b)

Arc Discharges

S. Yatom, A. Khrabry, J. Mitrani, A. Khodak, I. Kaganovich, V. Vekselman, B. Stratton and Y. Raitses "Synthesis of nanoparticles in carbon arc: measurements and modeling", MRS. Comm. (2018)





M. Kundrapu, M. Keidar, Numerical simulation of carbon arc discharge for nanoparticle synthesis, Physics of Plasma, Vol. 19, No. 073510, 2012

<u>Important quantities for **every** flow:</u>

- Temperature
- Density
- Flow velocity

WHAT EVEN IS TEMPERATURE????

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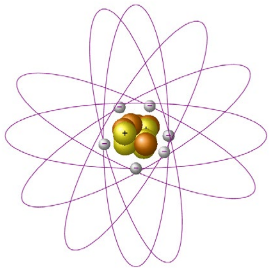
WHY CARE ABOUT FLOW METROLOGY?

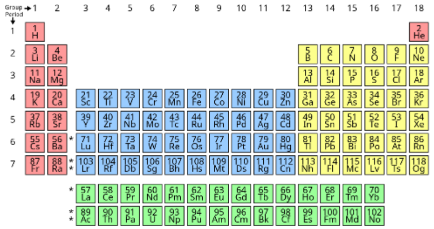
<u>Ideal characteristics for **every** diagnostic</u>:

✓ Non-perturbative

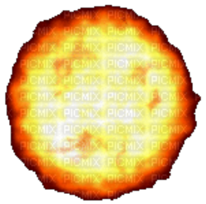


✓ Single shot

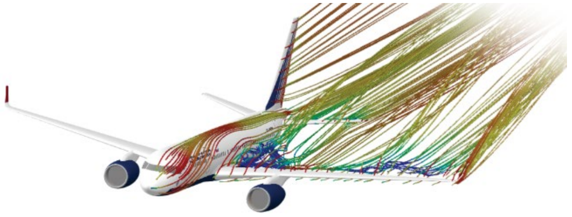
✓ Works with all species



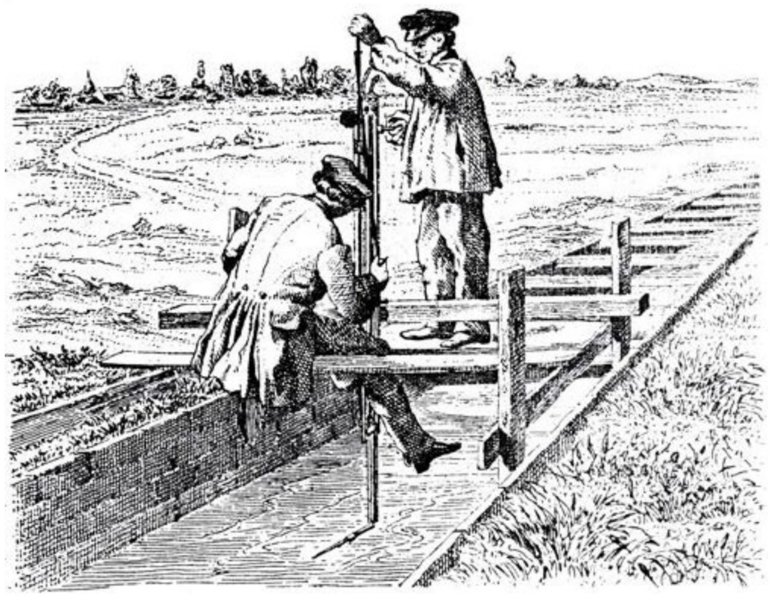
✓ Works *in-situ*, in a multitude of environments (e.g. wind-tunnels/fire/plasma)



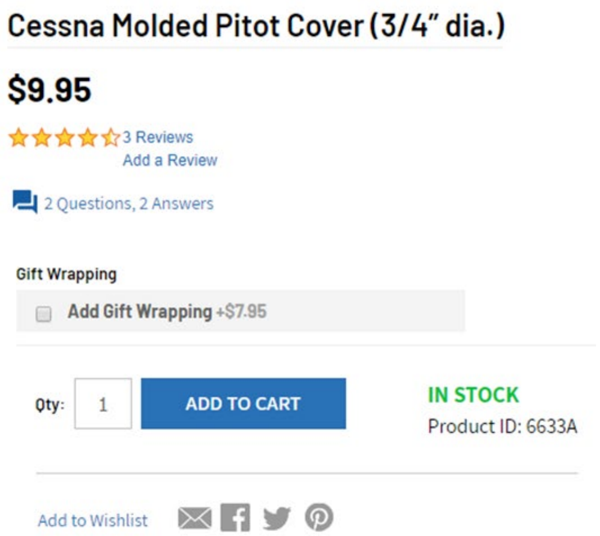
✓ Spatially maps out as much of the flow field as possible



WHY CARE ABOUT FLOW METROLOGY?
The state-of-the-art



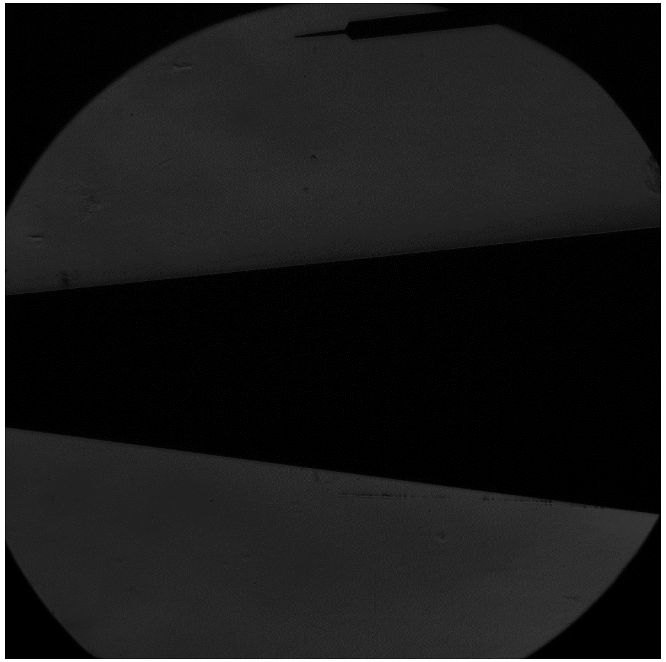
Measurement of flow velocity with the Pitot tube, improved by M. H. Darcy (Recherches hydrauliques, entreprises par M. H. Darcy, continuées par M. H. Bazin. Extract from volume XIX of Mémoires présentés par divers savants à L'institut Impérial de France, 1st part (501 p., 28 pl.): Recherches expérimentales sur l'écoulement de l'eau dans les canaux découverts. Imprimerie impériale, Paris 1865, planche IV).



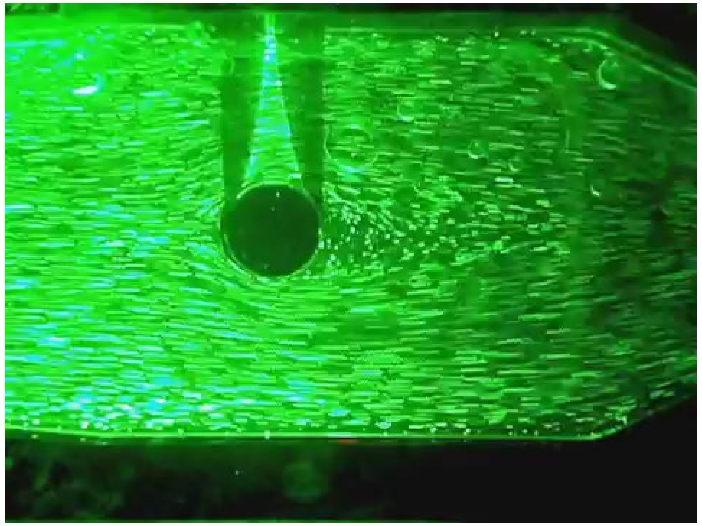
WHY CARE ABOUT FLOW METROLOGY?

The state-of-the-art

Hot Wire

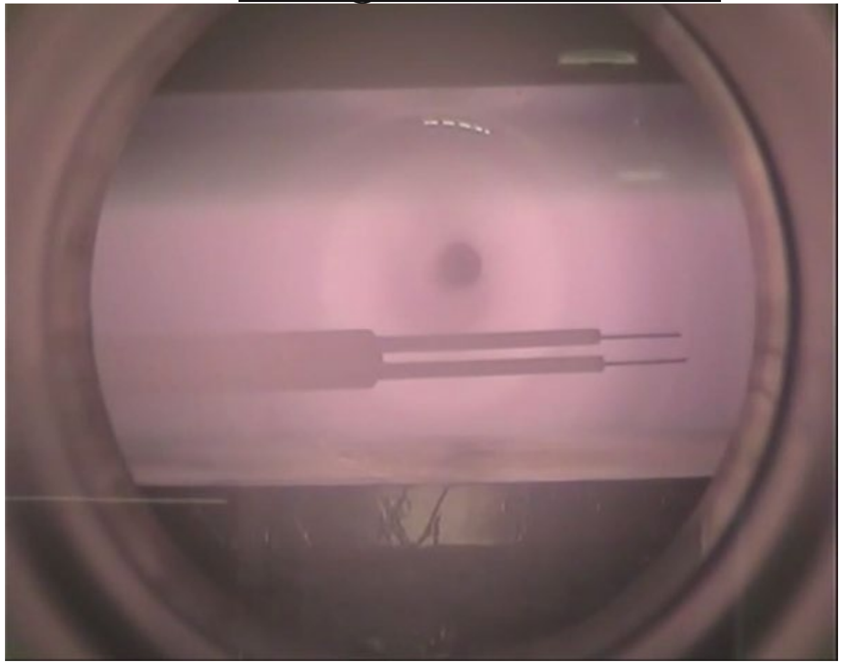


Particle Image Velocimetry (PIV)



Plasma & Plasma flow diagnostics:

Langmuir Probes



Taken from https://www.youtube.com/watch?v=6uZfcE80DDQ

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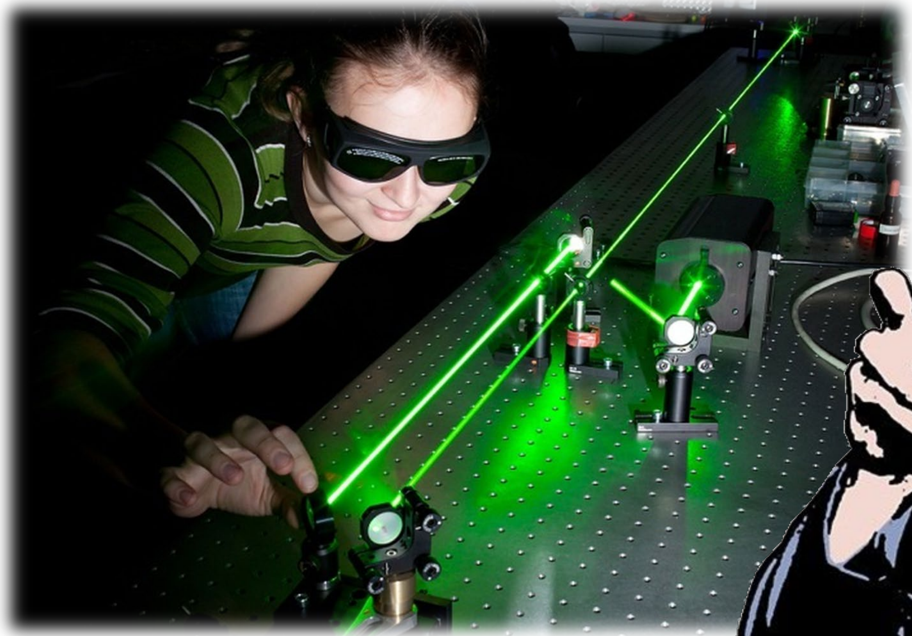
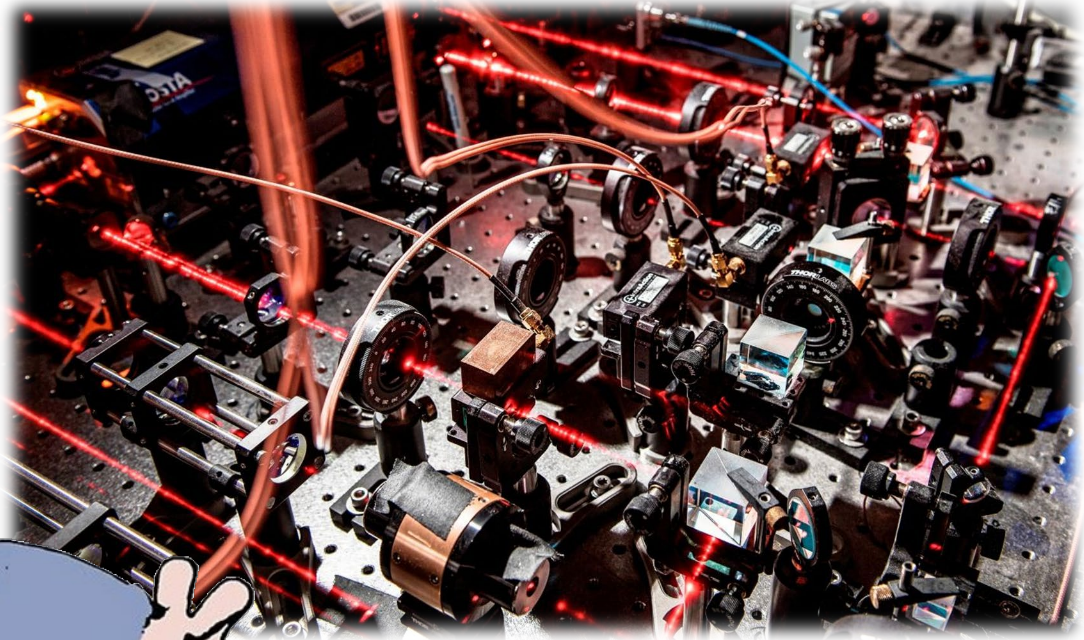
WHY CARE ABOUT FLOW METROLOGY?

The solution….

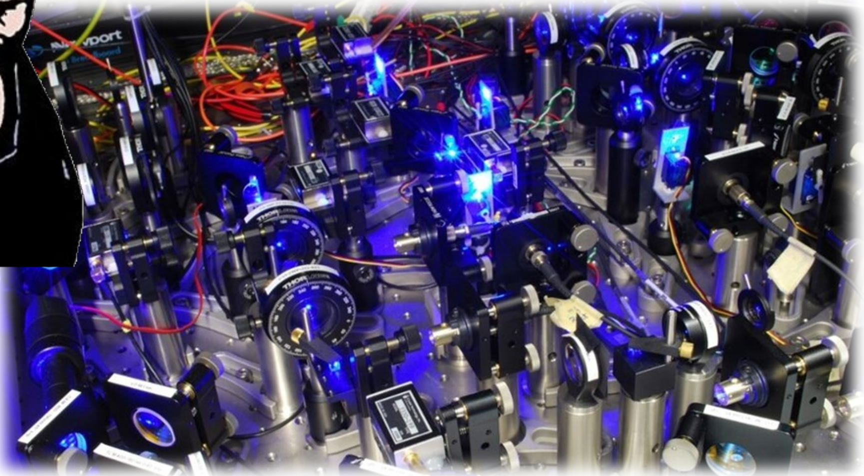


https://forbes.kz/process/science/5_awesome_things_s
do_with_lasers/

“LASERS”



https://www.eurekalert.org/multimedia/697040



https://phys.org/news/2021-11-laser-cooling-quantum-gases.html

Light
Amplification
(by) Stimulated
Emission
(of) Radiation

UK Atomic Energy Authority

FUSION | ROBOTICS | MATERIALS | SKILLS | PROTOTYPE

CCFE
CULHAM CENTRE FOR FUSION ENERGY

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Fifty years ago, five scientists from Culham Laboratory made a trip to the Soviet Union that was to prove a pivotal moment in the quest for fusion energy.

Their journey to Moscow's Kurchatov Institute in 1969 confirmed the impressive results of the Soviets' T3 experiment – a relatively new type of fusion device known as a 'tokamak'. The Culham team measured plasma temperatures of around 10 million degrees C at T3; far in excess of any previous fusion machine and a big step towards the conditions needed for fusion power. The emergence of the tokamak was a huge boost for fusion research and opened the way to machines like JET, ITER and a viable route to future power plants.

These days, international collaboration in science is taken for granted. But in the late 1960s, the Cold War was at its peak and mutual suspicion between East and West made the Culham scientists' mission an almost unprecedented breach of the 'Iron Curtain'.

To mark the 50th anniversary of the publication of the T3 results in Nature, we spoke to Dr Mike Forrest, who was on the team that went to Moscow. Mike helped to develop the 'Thomson scattering' laser techniques that made the measurements possible and that are still a key diagnostic for today's tokamaks. Mike recalls the pioneering early use of lasers, the top-level clearance needed for the trip and how UK and Soviet scientists bonded over a shared love of tea as they made a breakthrough that still resonates today.

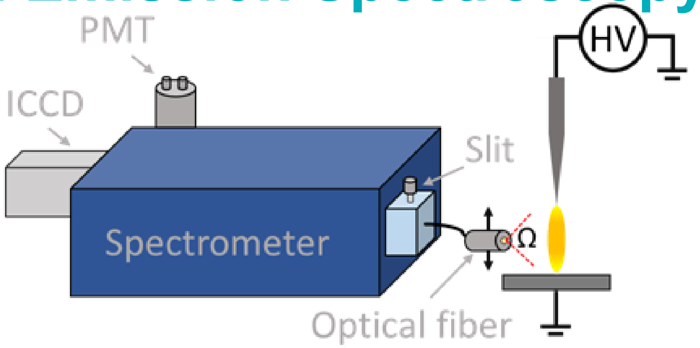
50 years on: The mission to Moscow that changed fusion research

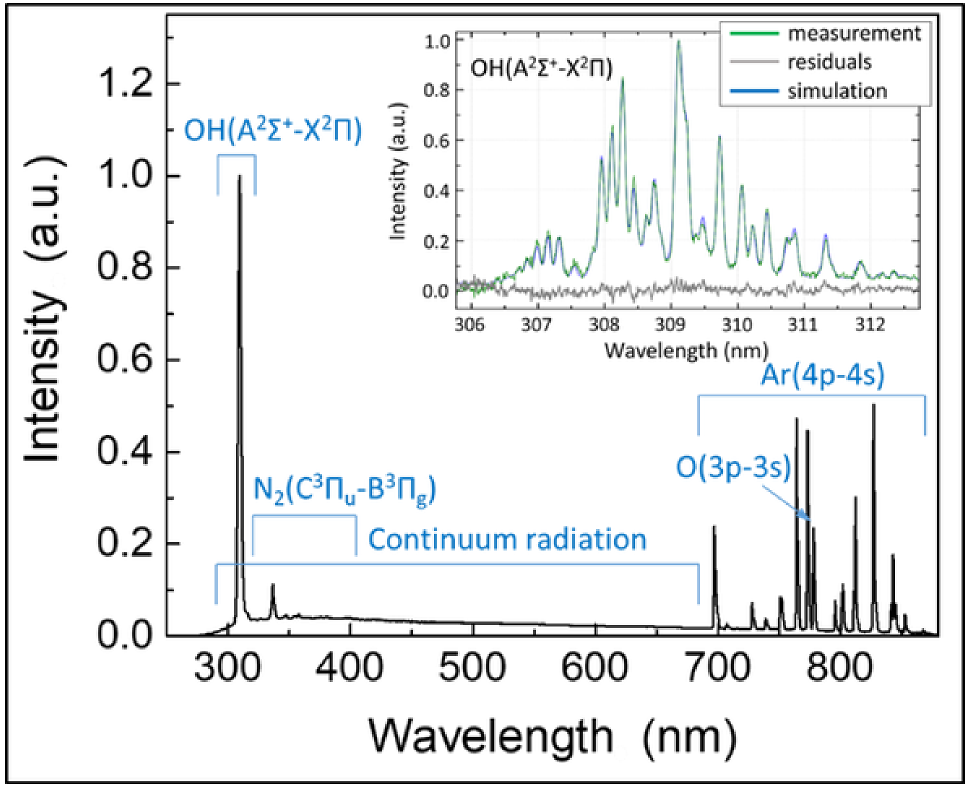
Watch Later | Share

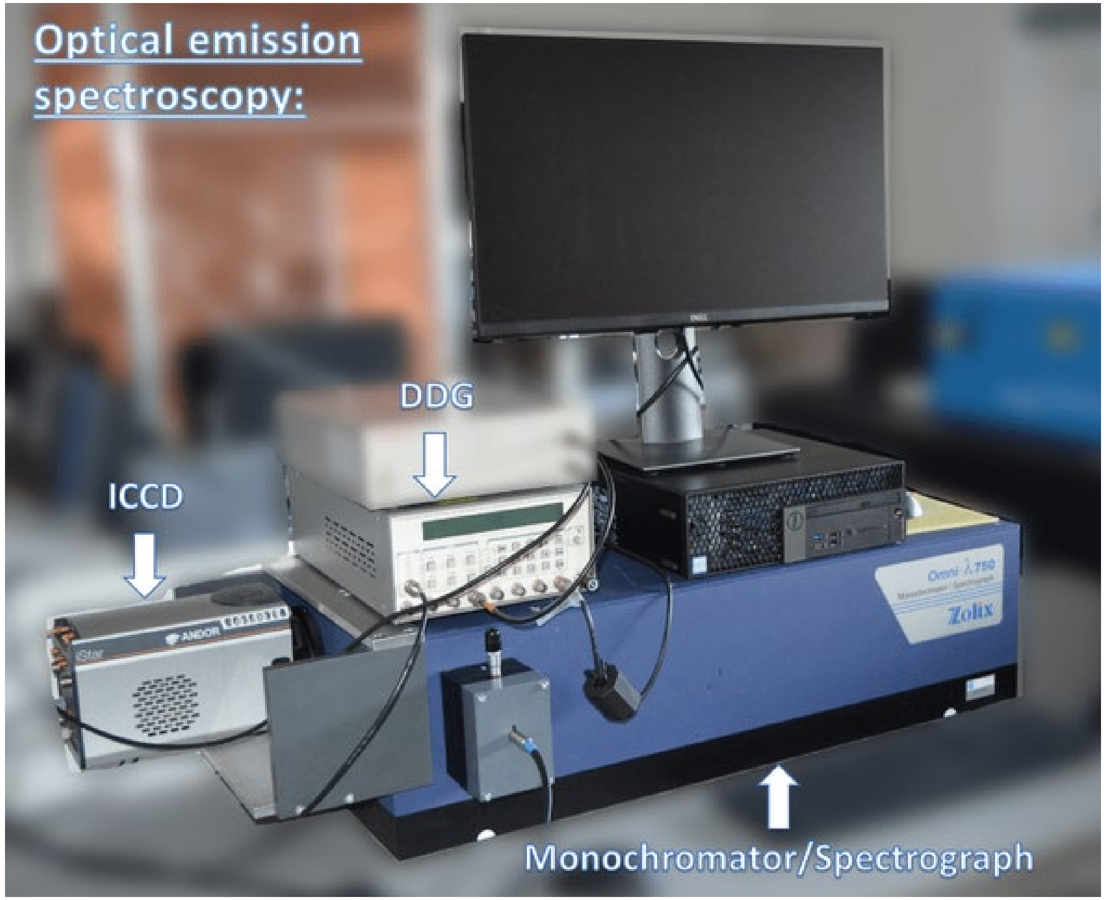
https://ccfe.ukaea.uk/mission-to-moscow-50-years-on/

WITHOUT LASERS....

Optical Emission Spectroscopy







https://www.ugent.be/ea/appliedphysics/en/research/plasma/plasmaphysicsandengineering.htm/plasmadiagnostics.htm

SO....WHAT IS TEMPERATURE?

Dictionary

Definitions from Oxford Languages · Learn more

temperature

noun

the degree or intensity of heat present in a substance or object, especially as expressed according to a comparative scale and shown by a thermometer or perceived by touch.
"at a temperature of 2°C"

- the degree of internal heat of a person's body.
  "I'll **take her temperature**"
- INFORMAL
  a body temperature above the normal.
  "he **was running a temperature**"

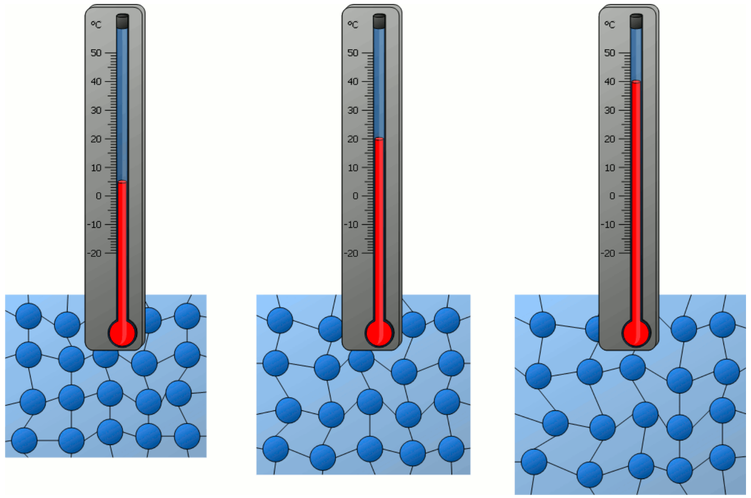
Wikipedia
https://en.wikipedia.org › wiki › Temperature

Temperature - Wikipedia

Temperature **is a physical quantity that expresses quantitatively the perceptions of hotness and coldness**. Temperature is measured with a thermometer.

Thermodynamic temperature · Atmospheric temperature · Scale of temperature

Temperature is a measure of the average kinetic energy per particle in a substance.



https://www.tec-science.com/thermodynamics/temperature/temperature-and-particle-motion/

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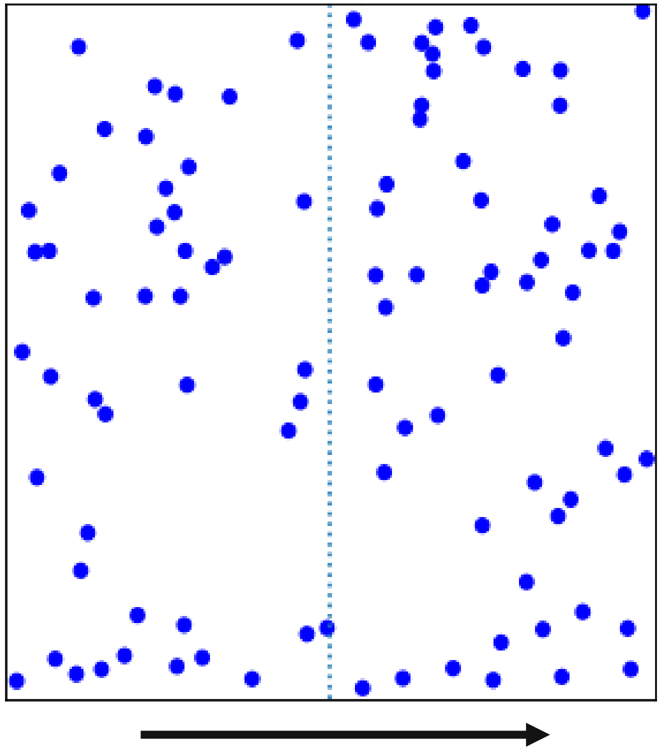
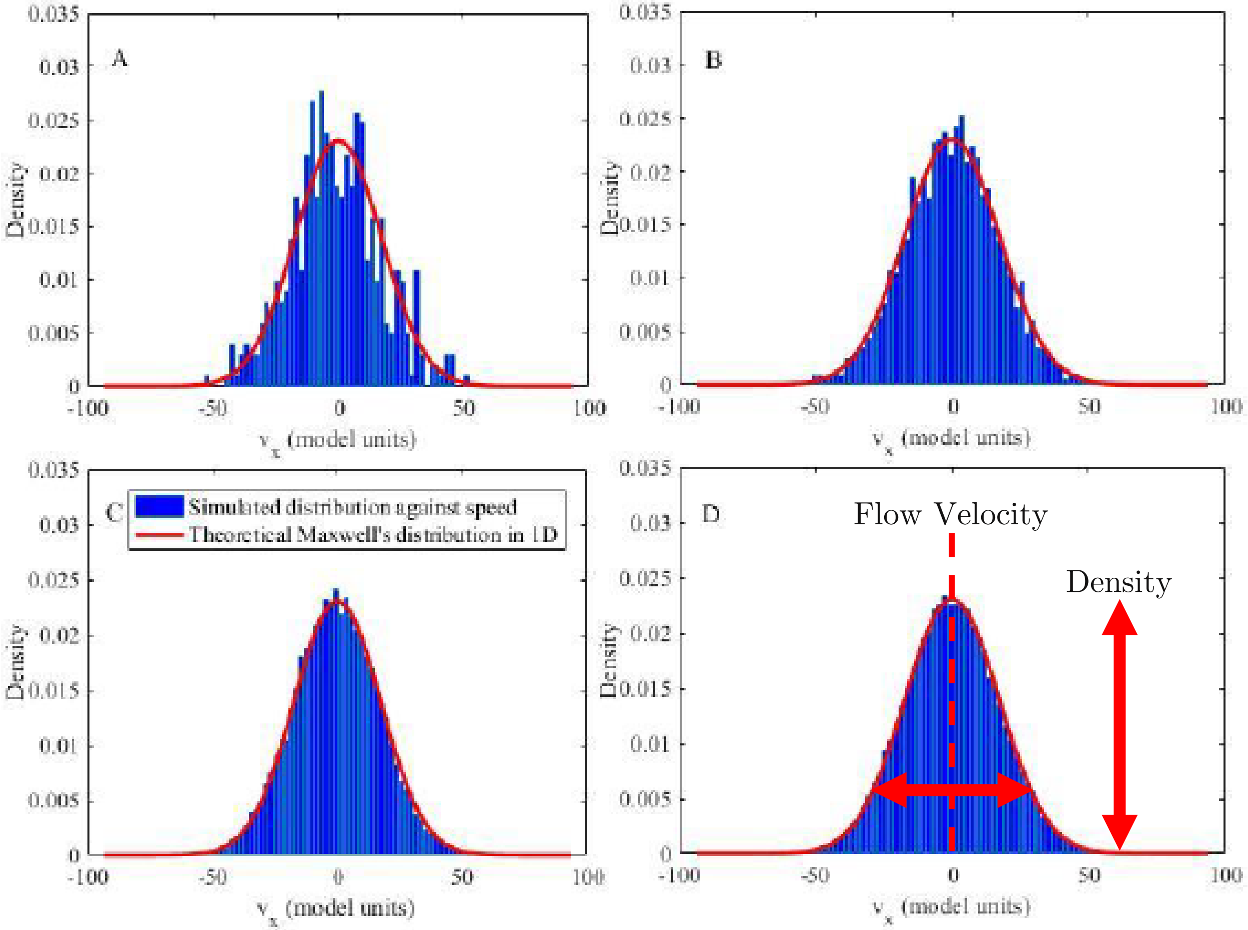
SO....WHAT IS TEMPERATURE?





https://stanford.edu/~jeffjar/statmech/week8a.html

doi.org/10.51724/ijpce.v13i2.142

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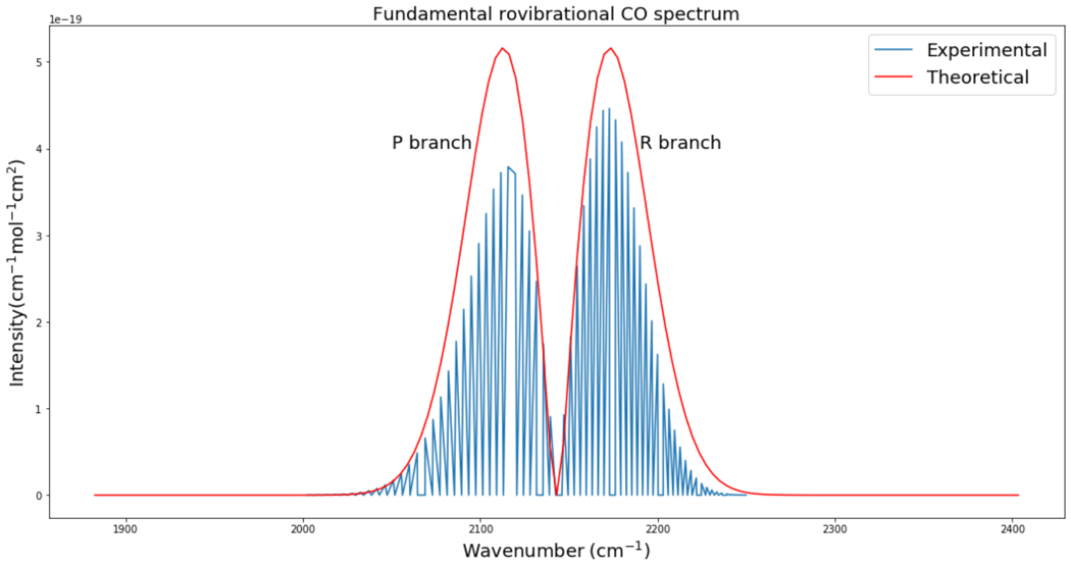
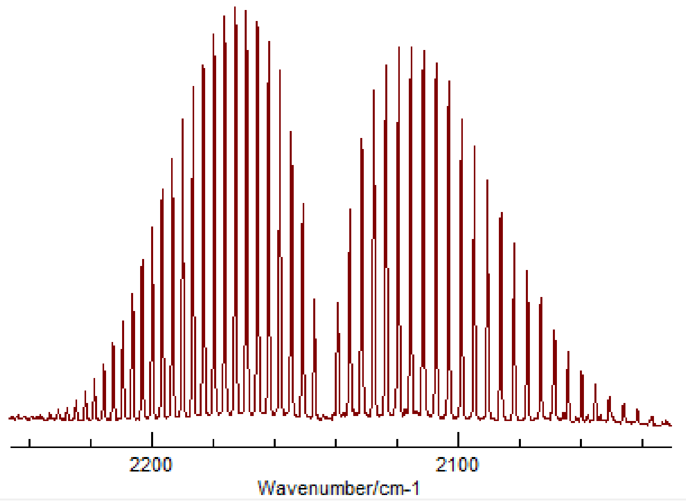
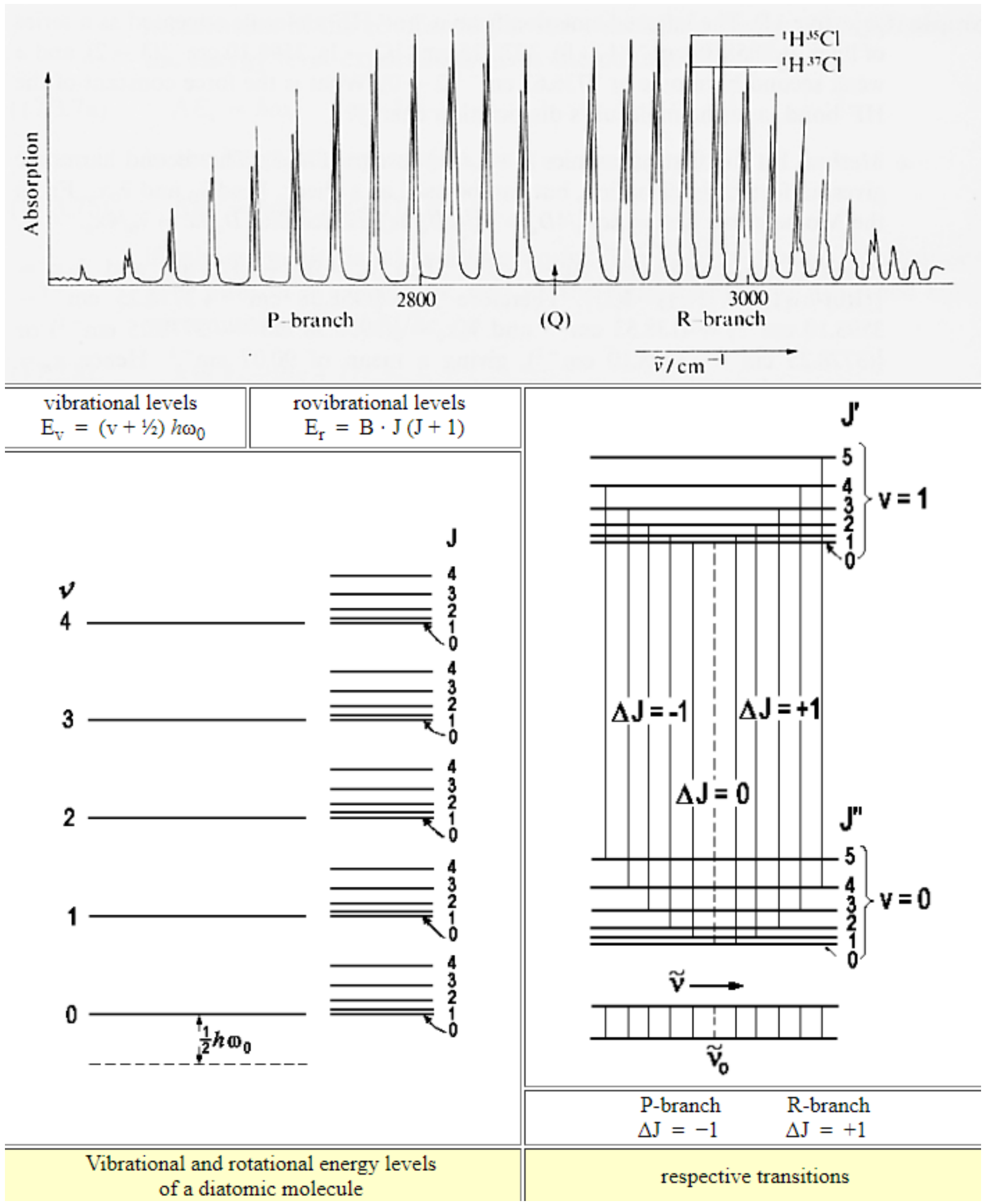
SO....WHAT IS TEMPERATURE?



https://pages.vassar.edu/magnes/2018/12/15/fundamental-rovibrational-spectrum-of-co/



https://commons.wikimedia.org/wiki/File:Carbon_monoxide_rotational-vibrational_spectrum.png



http://www.pci.tu-bs.de/aggericke/PC4e/Kap_III/Rot-Vib-Spektren.htm

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SO....WHAT IS TEMPERATURE?

When:

$$\underbrace{\textbf{Translational} = \textbf{Vibrational} = \textbf{Rotational} = \textbf{Electronic}}_{\textbf{Thermodynamic Equilibrium}}$$

SO….WHAT IS TEMPERATURE?

When:

$$\underbrace{\textbf{Translational} \neq \textbf{Vibrational} \neq \textbf{Rotational} \neq \textbf{Electronic}}$$

Thermodynamic Non-Equilibrium

SO....WHAT IS TEMPERATURE?

Temperature is defined through a Maxwellian distribution function...

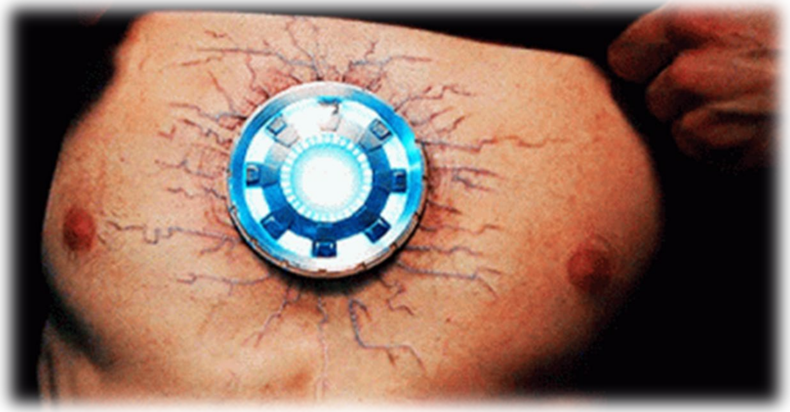
What if our particles do **not** follow a Maxwellian distribution function?

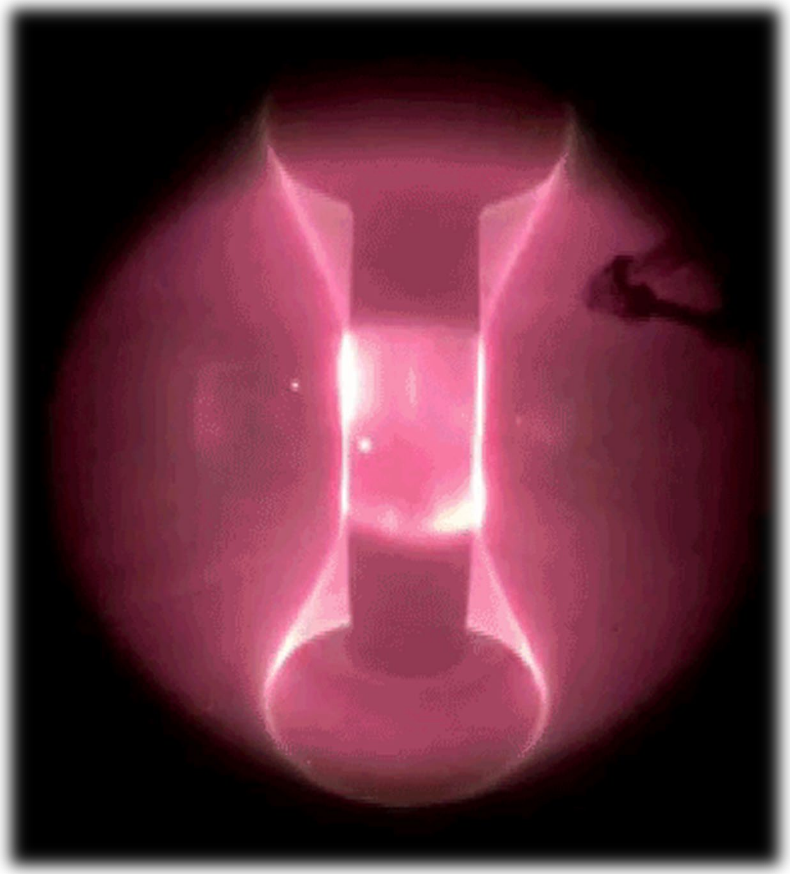
How is temperature defined then???

LUXEMBOURG INSTITUTE OF SCIENCE AND TECHNOLOGY LIST

WHAT DO WE WANT TO MEASURE IN A PLASMA???

Xenon (@15 Torr)/ Gerakis lab





Footage from the inside of the NSTX-U during a run. Image Courtesy of PPPL.

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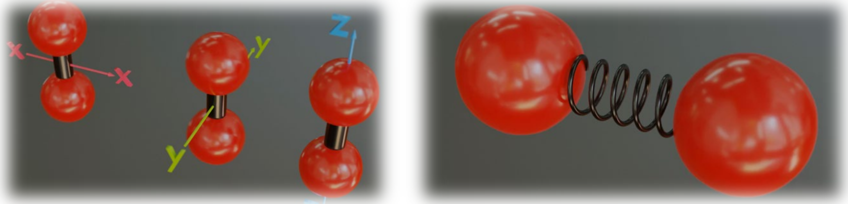
WHAT DO WE WANT TO MEASURE IN A PLASMA???

- **Electrons**: Density, Temperature, Flow velocity
- **Atoms (neutrals)**: Density, Temperature, Flow velocity
- **Atomic ions**: Density, Temperature, Flow velocity
- **Molecules (neutrals)**: Density, Temperature, Flow velocity



Rotational Temperature

Vibrational Temperature

- **Molecular ions**: Density, Temperature, Flow velocity

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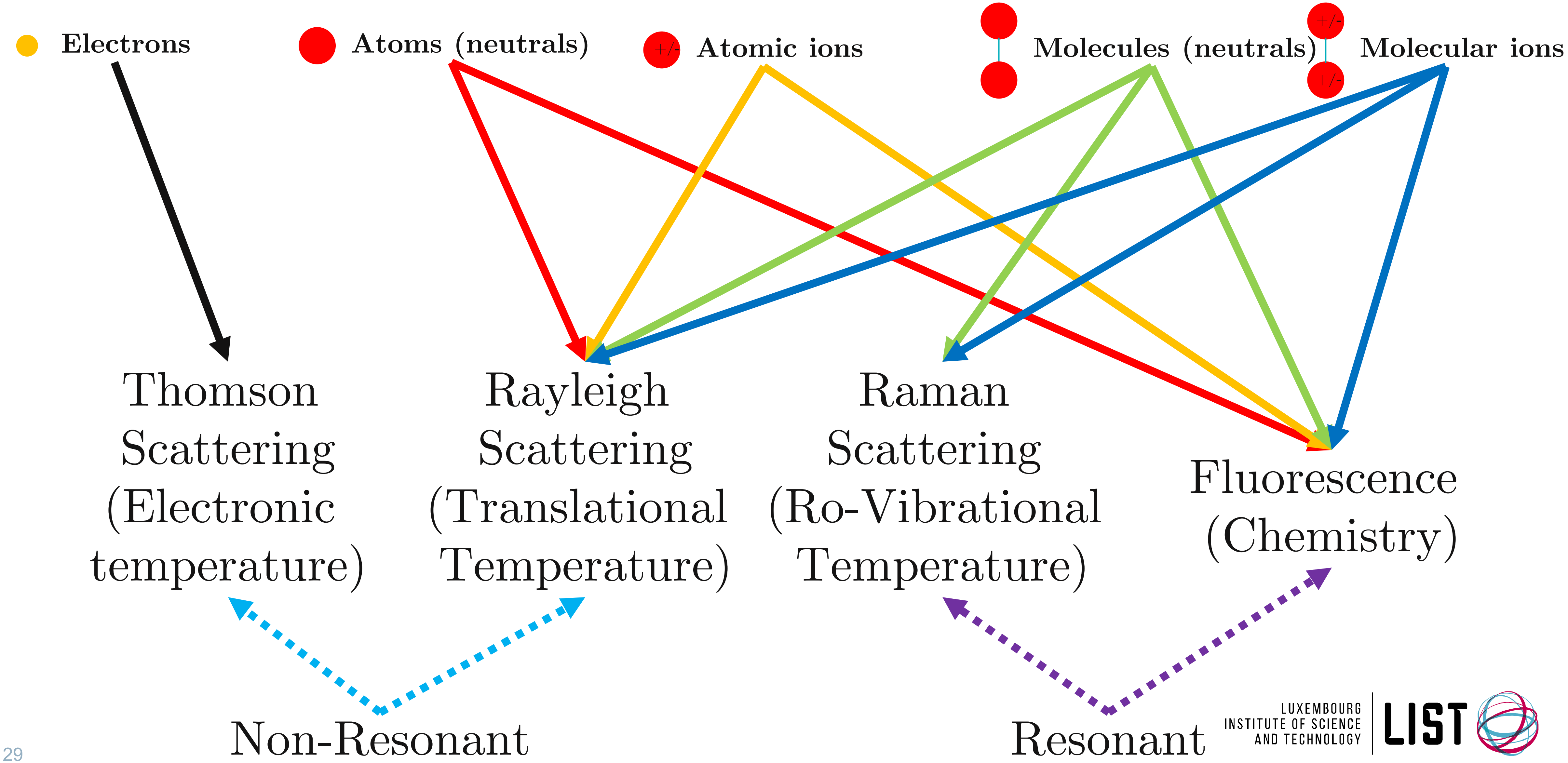
HOW DO WE MEASURE THINGS IN A PLASMA???

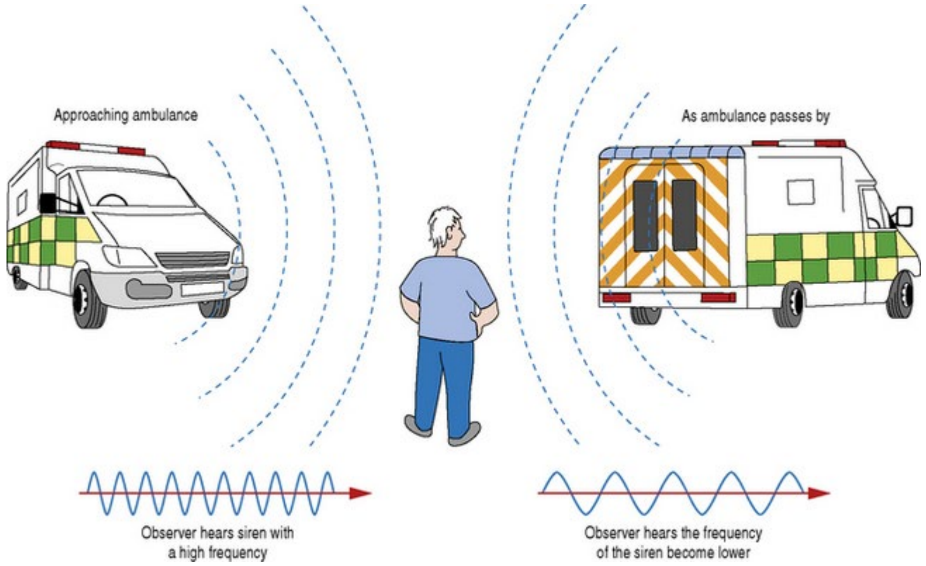


HOW DO WE MEASURE THINGS IN A PLASMA???

LINEAR REGIME

THOMSON SCATTERING

- Electrons

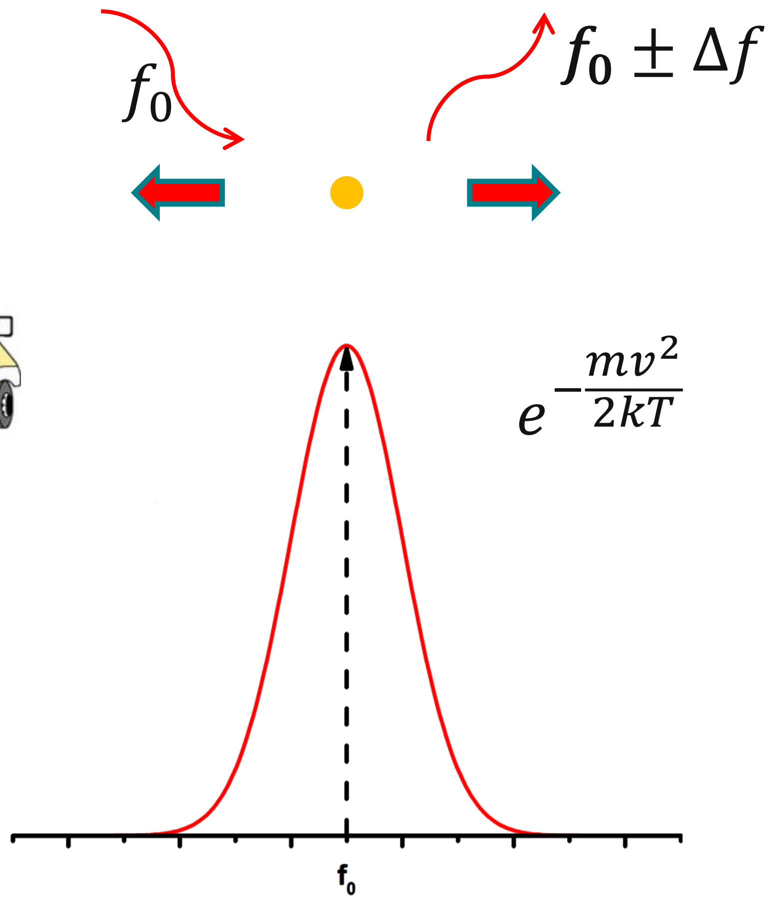


https://radiologykey.com/physical-principles-of-doppler-ultrasound/

f_0

$f_0 \pm \Delta f$

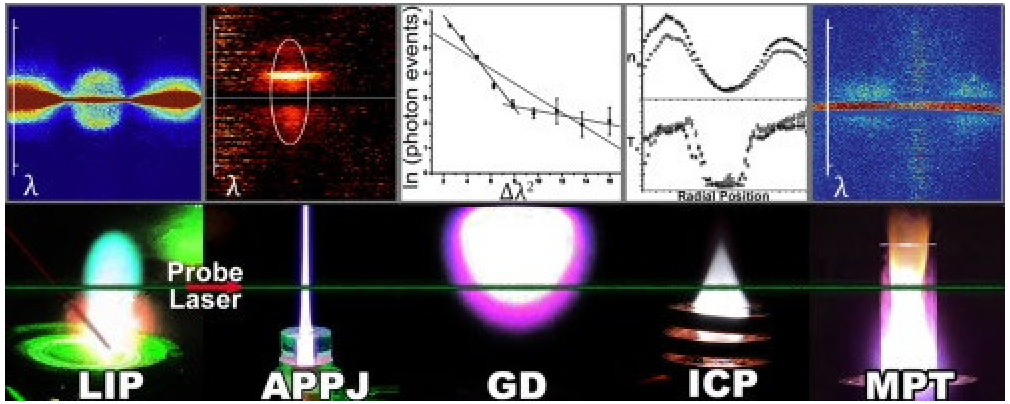
$e^{-\frac{mv^2}{2kT}}$



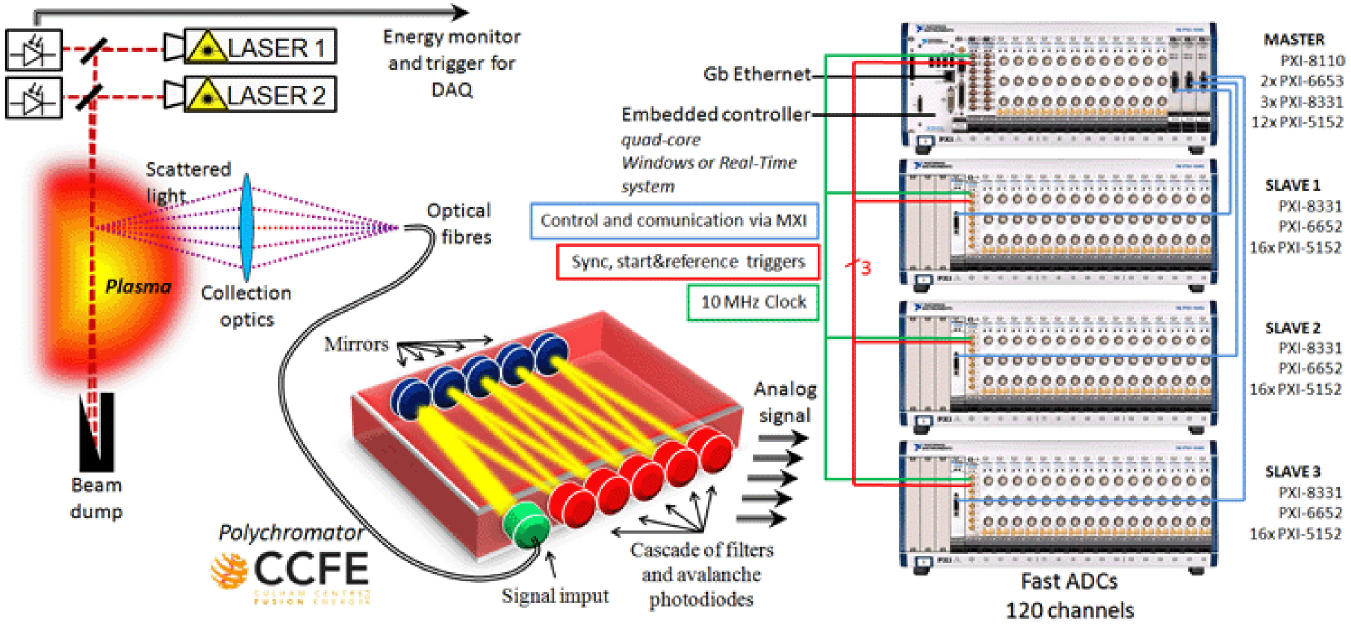
Electronic temperature

THOMSON SCATTERING

- Electrons



doi.org/10.1016/j.sab.2020.106045



https://www.ipp.cas.cz/vedecka_struktura_ufp/tokamak/COMPASS/diagnostics/spektroskopicke-diagnostiky/thomsonuv-rozptyl.html

Typical cross-sections: 10^{-29} - 10^{-30} m^{-2}

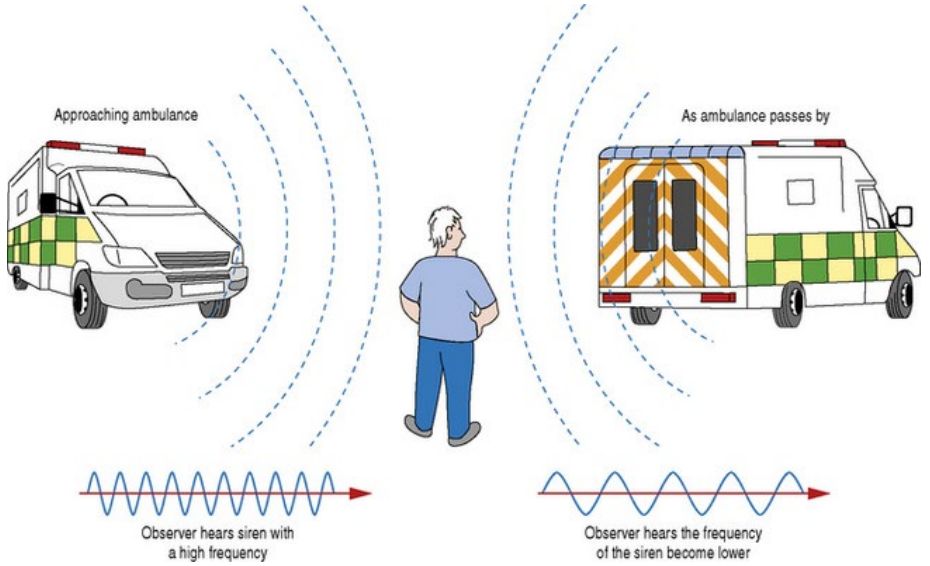
RAYLEIGH SCATTERING

Atoms (neutrals)

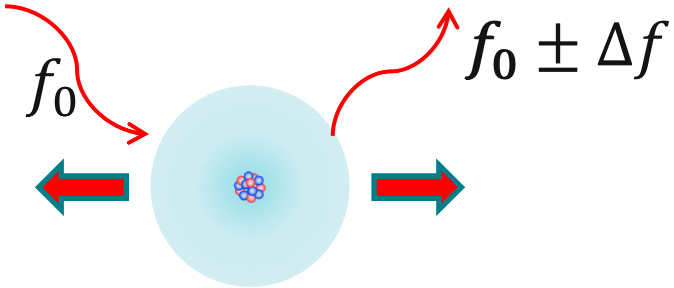
Atomic ions

Molecules (neutrals)

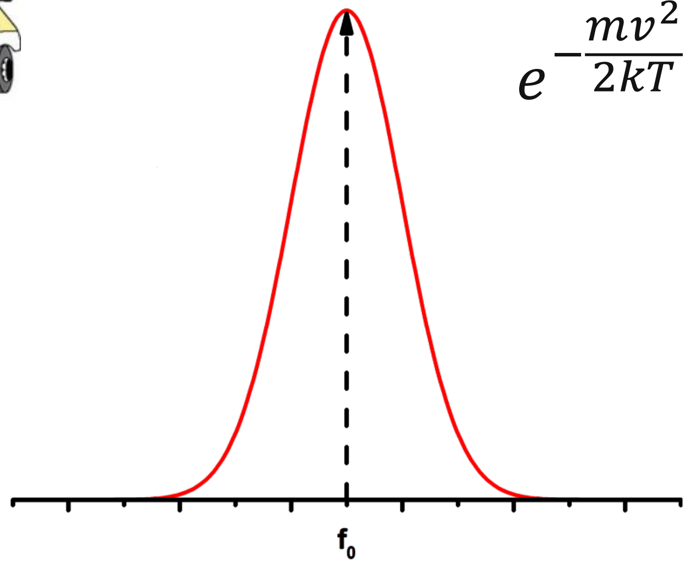
Molecular ions



https://radiologykey.com/physical-principles-of-doppler-ultrasound/



$$e^{-\frac{mv^2}{2kT}}$$



Translational temperature

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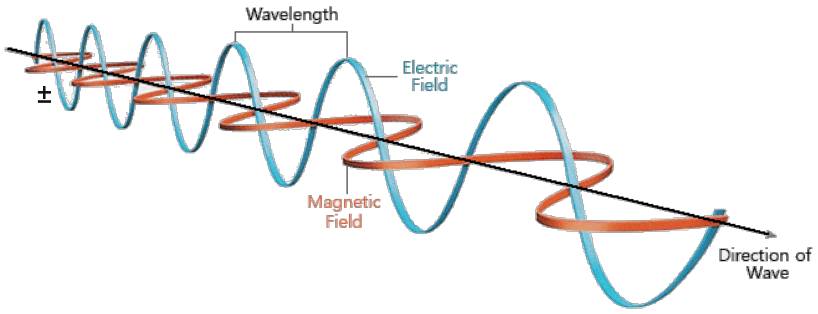
RAYLEIGH SCATTERING

Atoms (neutrals)

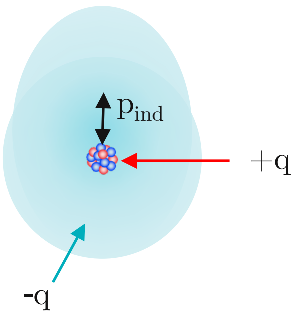
Atomic ions

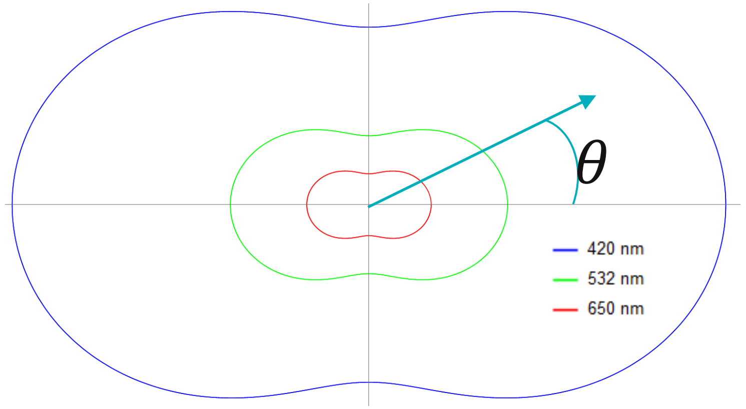
Molecules (neutrals)

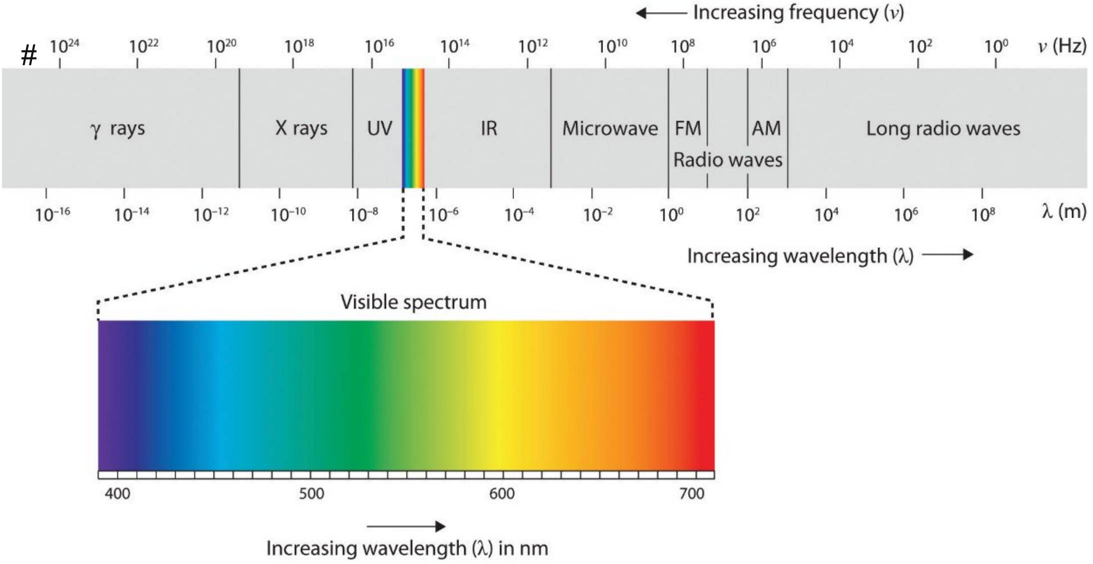
Molecular ions



$$p_{ind} = \alpha \cdot E$$







$$I = I_0 \frac{8\pi^4 N\alpha^2}{\lambda^4 R^2}(1 + cos^2\theta)$$

N=# of scatterers

α= polarizability

R= distance from scatterer

± Image taken from http://emf.kca.kr/publish/Images/elec_business/img_elec_business_business01.gif (accessed 4/22/2018)

Image taken from https://support.precisionmapper.com/support/solutions/articles/6000185574-multispectral-sensors

RAYLEIGH SCATTERING

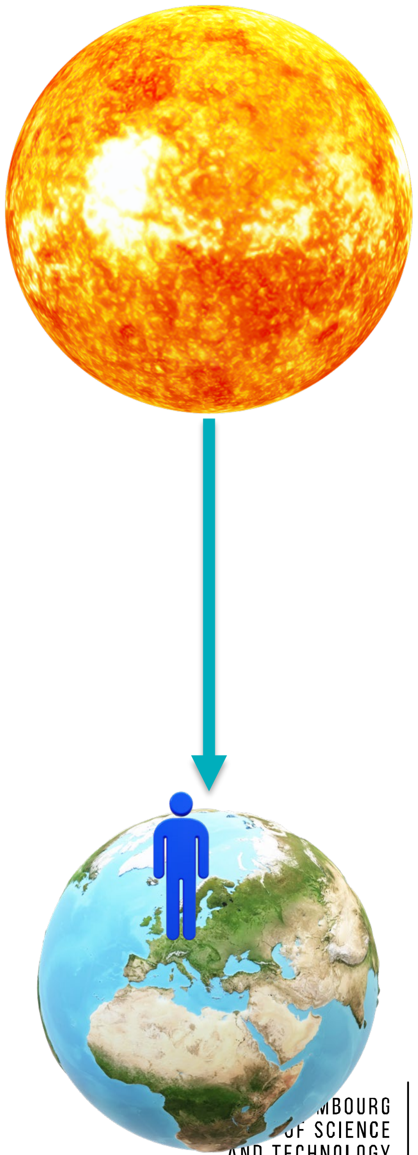
Atoms (neutrals)

Atomic ions

Molecules (neutrals)

Molecular ions

Image taken from https://ceej.travellerspoint.com/157/



RAYLEIGH SCATTERING

Atoms (neutrals)

Atomic ions

Molecules (neutrals)

Molecular ions

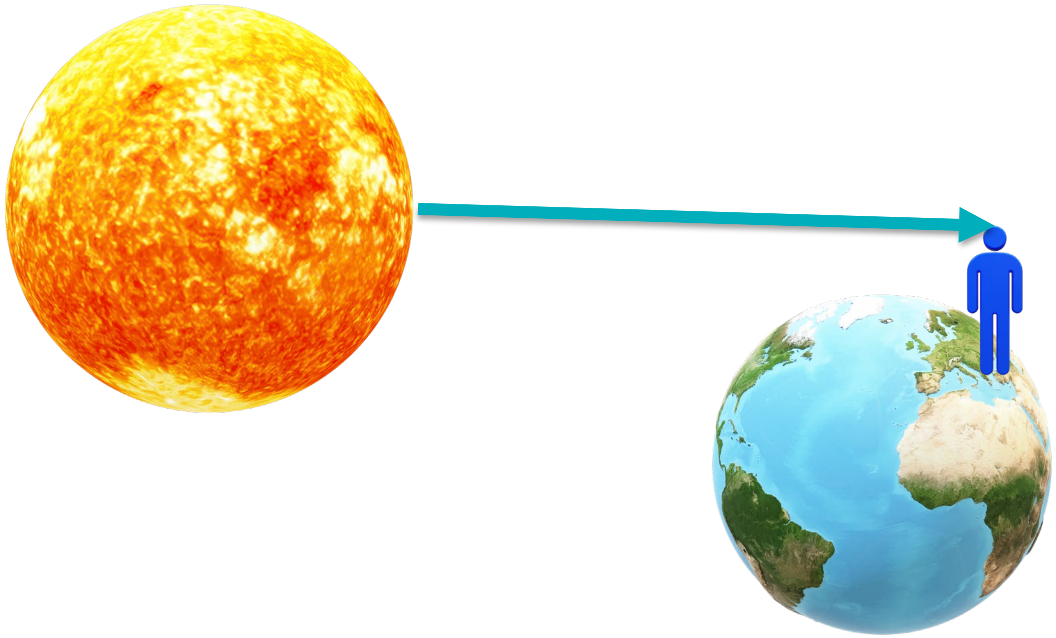
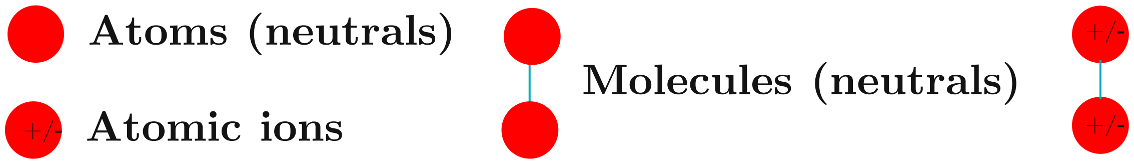


Image taken by AG somewhere in the Aegean

RAYLEIGH SCATTERING

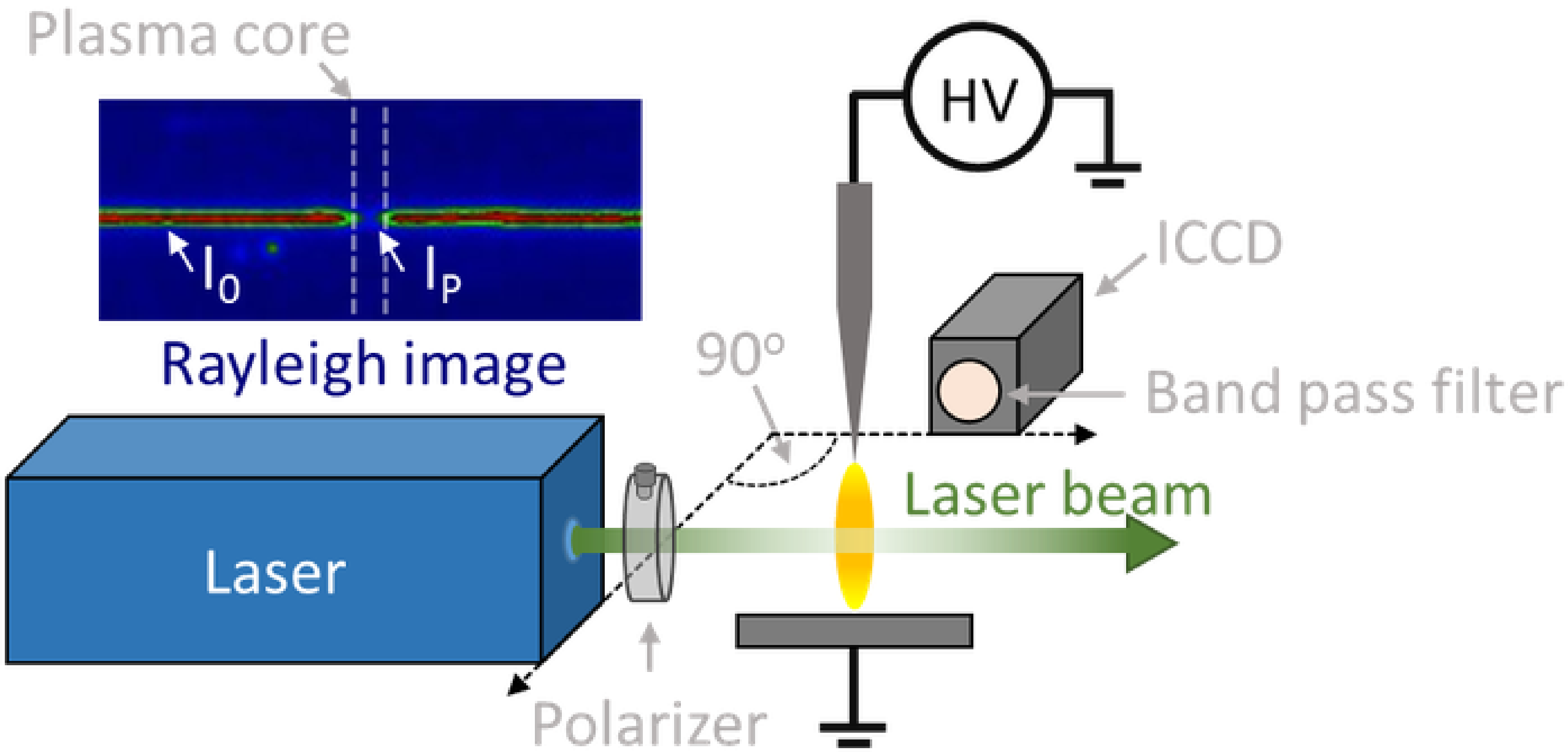


Atoms (neutrals)

Atomic ions

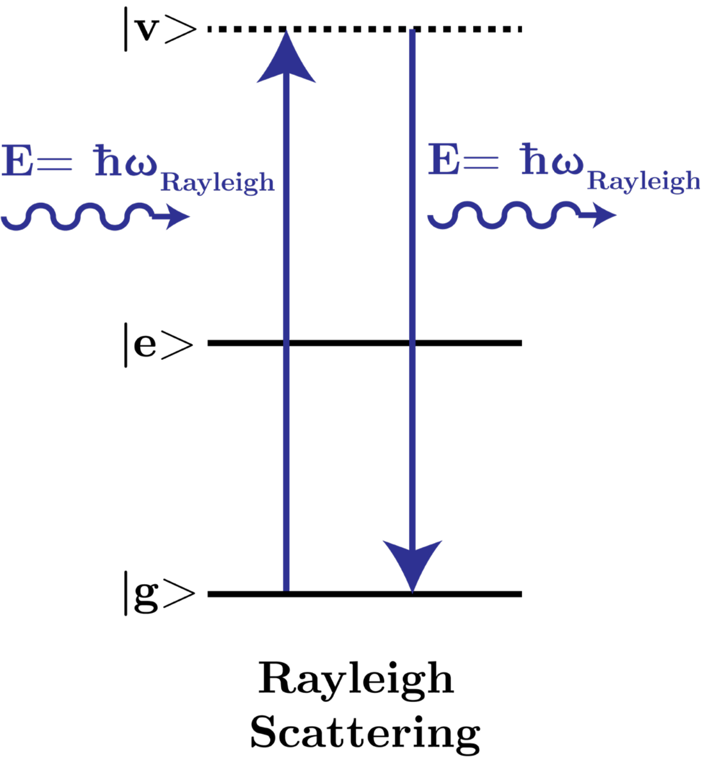
Molecules (neutrals)

Molecular ions



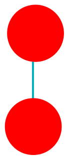
https://www.ugent.be/ea/appliedphysics/en/research/plasma/plasmaphysicsandengineering.htm/plasmadiagnostics.htm

RESONANT & NON-RESONANT APPROACHES

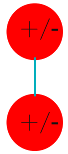


(elastic) (inelastic) (inelastic)

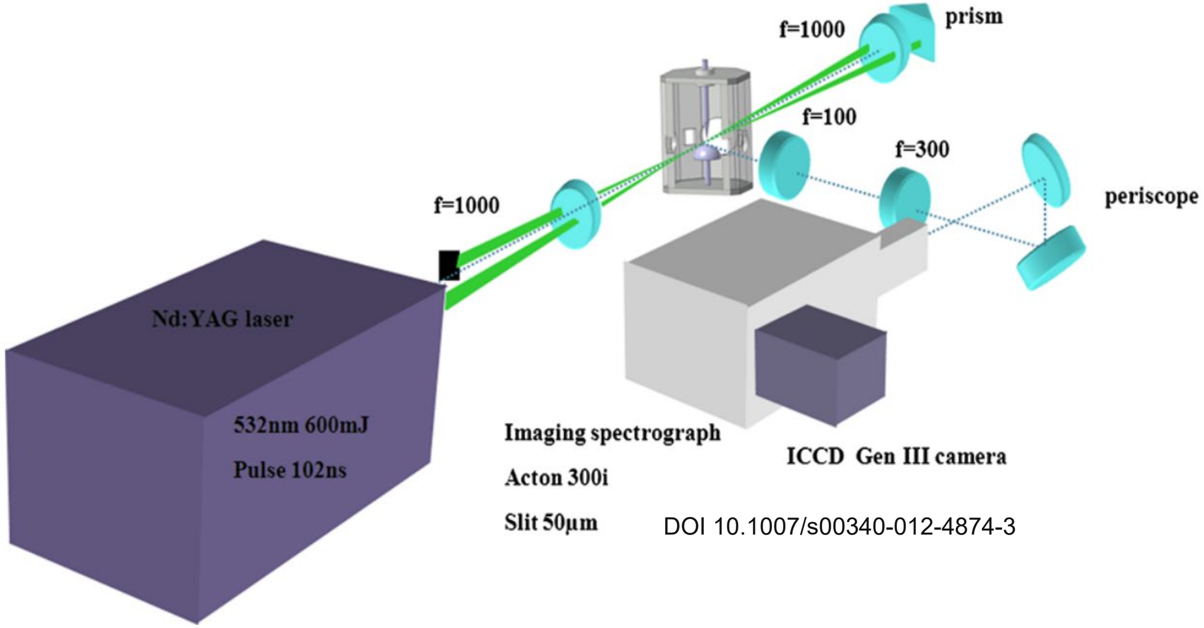
SPONTANEOUS RAMAN SCATTERING



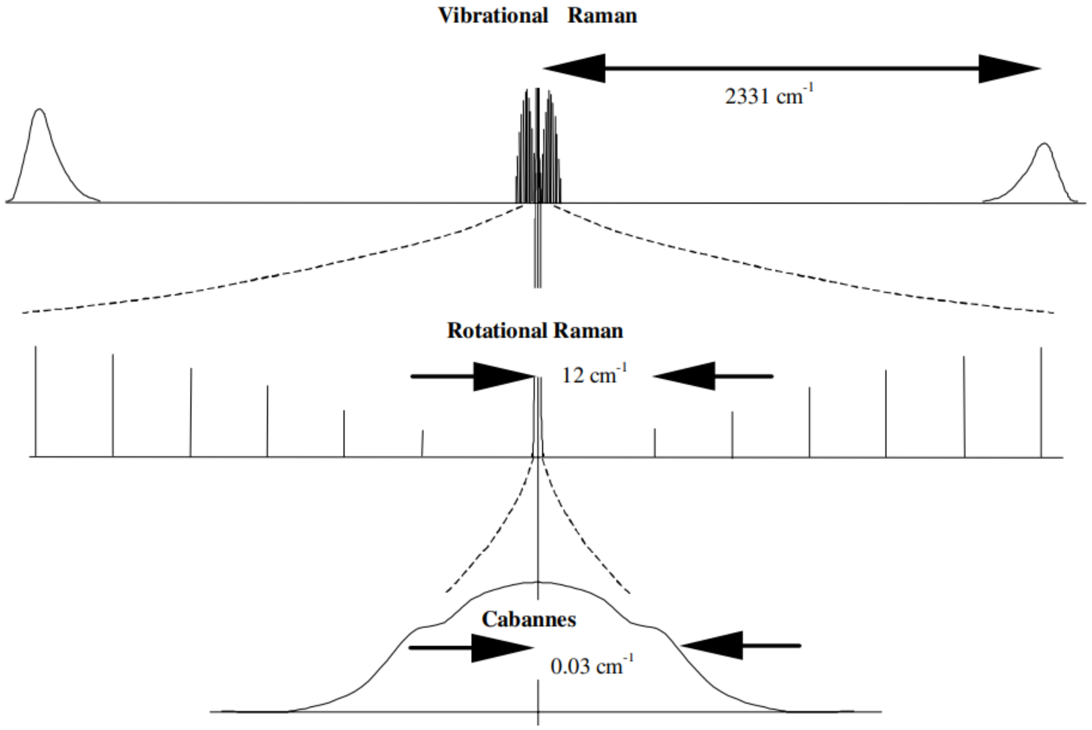
Molecules (neutrals)



Molecular ions



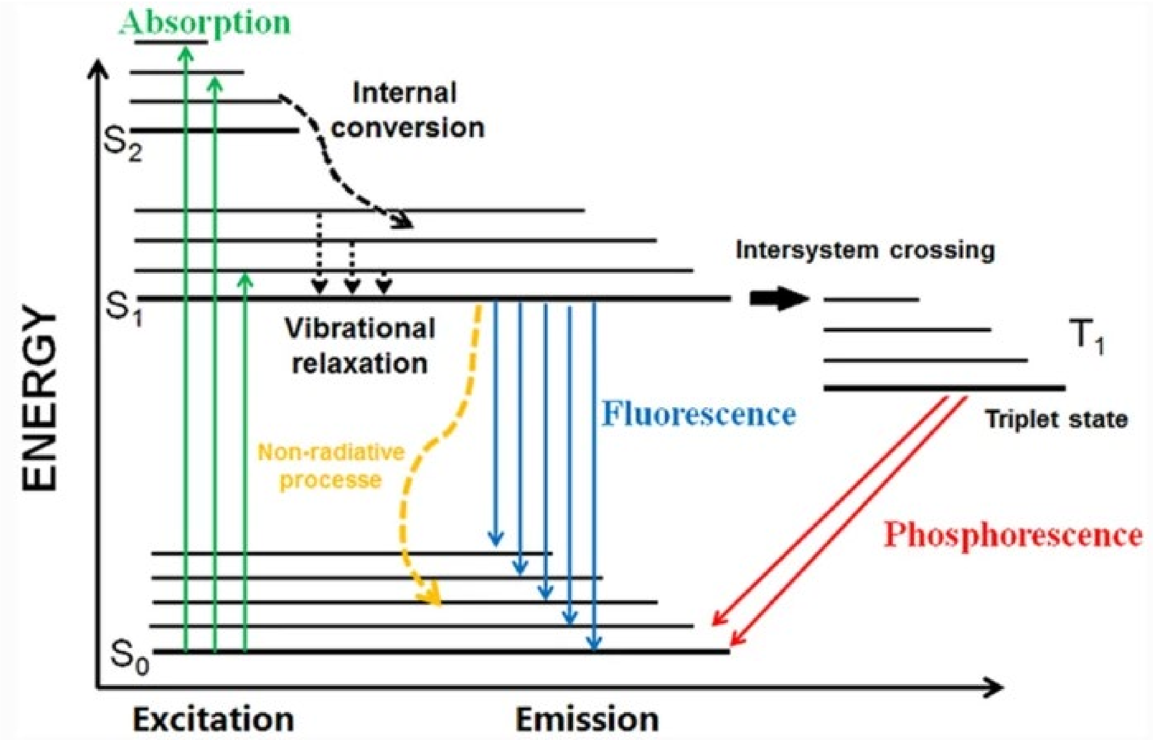
DOI 10.1007/s00340-012-4874-3



DOI 10.1088/0957-0233/12/5/201

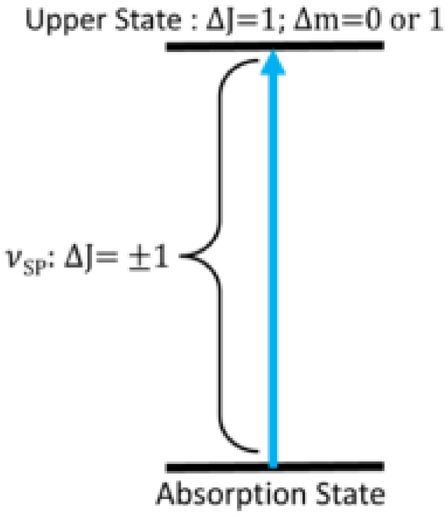
LASER INDUCED FLUORESCENCE

- Atoms (neutrals)
- +/- Atomic ions
- Molecules (neutrals)
- +/- +/- Molecular ions



https://link.springer.com/article/10.1007/s41104-021-00092-3#ref-CR12

https://researchrepository.wvu.edu/cgi/viewcontent.cgi?article=6578&context=etd



LIF

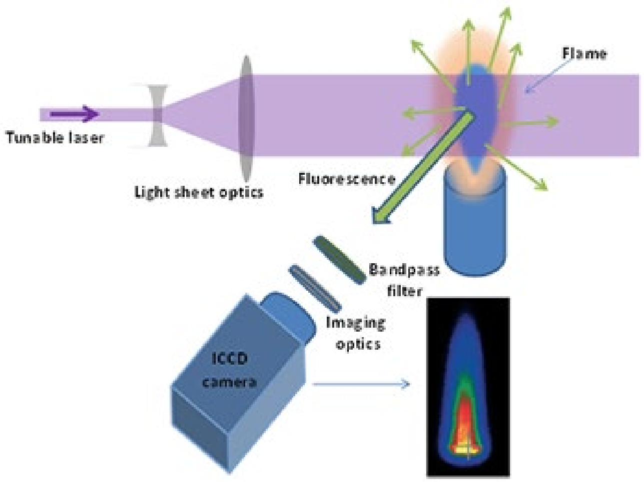
LASER INDUCED FLUORESCENCE

Atoms (neutrals)

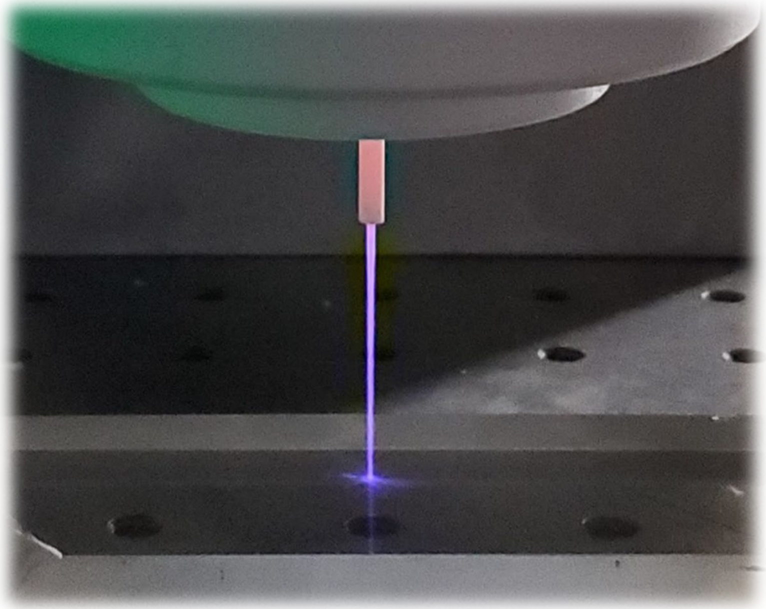
Atomic ions

Molecules (neutrals)

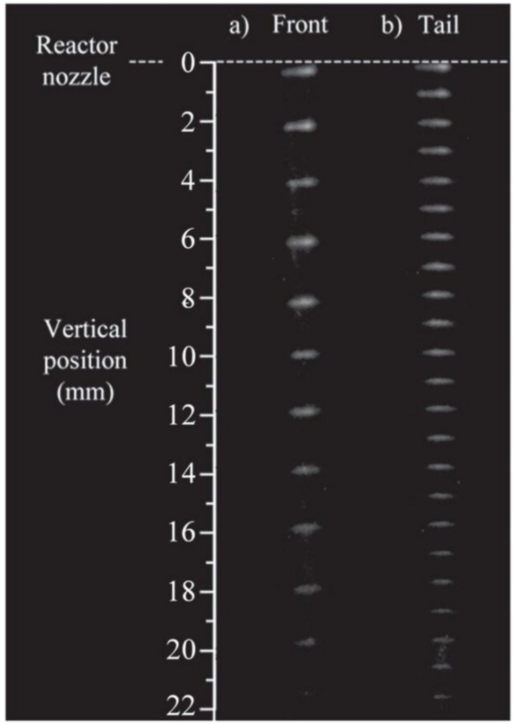
Molecular ions



https://andor.oxinst.com/learning/view/article/planar-laser-induced-fluorescence-as-a-plasma-diagnostic



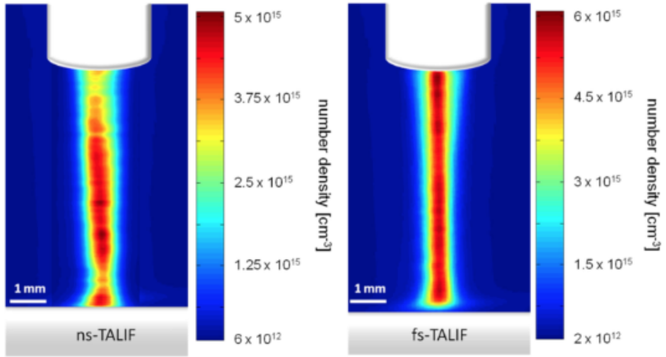
CAPPS/Gerakis lab



•10.1088/1402-4896/acc906

2D TALIF Image of O atom: ns vs fs comparison

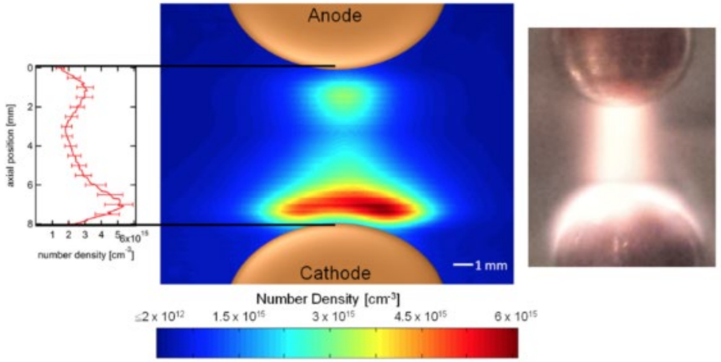
[Schmidt et al., Plasma Sources Science and Technology **26** (5), 055004 (2017)]



Comparison of 2D image of the O atom employing **(a)** ns-TALIF and **(b)** fs-TALIF for the same discharge in 2%-O2**/**He-mixture APPJ. Minimum detection limit of fs-TALIF is factor-of-three lower than ns-TALIF.

H-atom plasma image

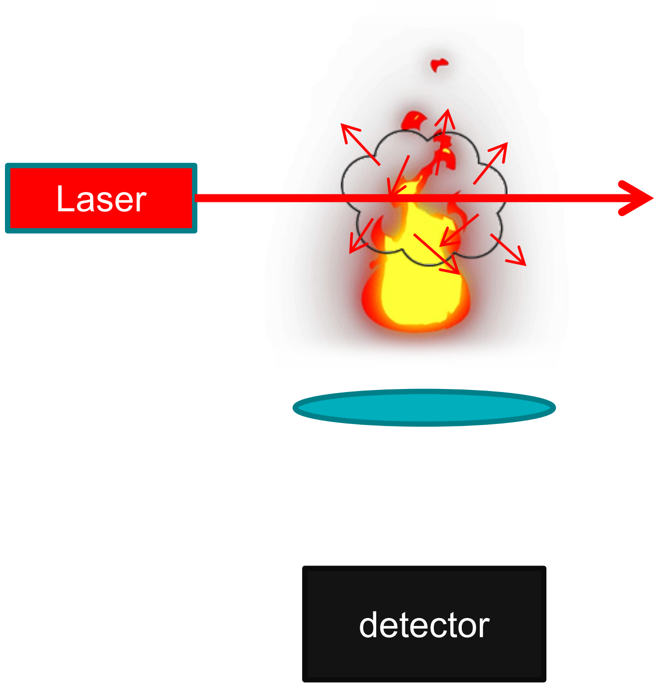
[Schmidt et al., Journal of Physics D: Applied Physics **50** (1), 015204 (2017)]



https://spectralenergies.com/research/electric-field-plasmas/plasma-imaging/

LINEAR PLASMA DIAGNOSTICS WORK GREAT!

Until….



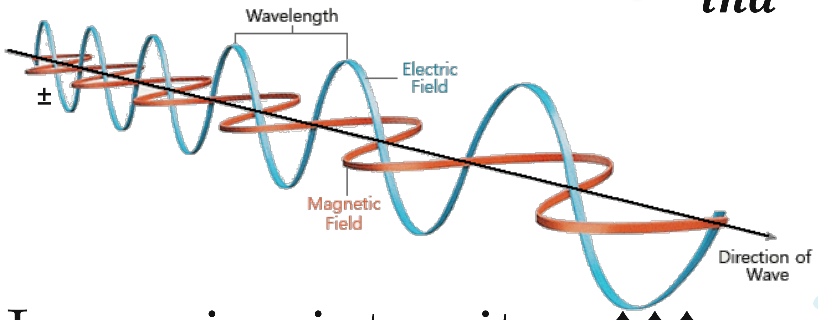
Other neutral and plasma flow laser diagnostics, resonant and non-resonant, e.g. vibrationally excited nitric oxide monitoring (VENOM), femtosecond laser electronic excitation tagging (FLEET) laser induced fluorescence (LIF), Coherent Anti-Stokes Raman Scattering (CARS) and others have also been demonstrated to measure flow velocity and/or temperature.

NON-LINEAR REGIME

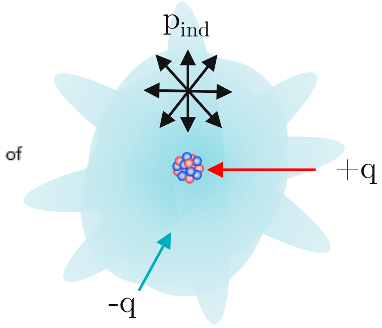
NON-LINEAR OPTICS

$$\boldsymbol{p}_{ind} = \alpha \cdot \boldsymbol{E}_{local} \quad \text{Microscopic}^{\dagger}$$

$$\boldsymbol{P}_{ind} = \varepsilon_0 \cdot \chi \cdot \boldsymbol{E} \quad \text{Macroscopic}$$



Increasing intensity...↑↑↑



As the light intensity increases, this *linear* model begins to breakdown. Instead, we now use a power series to model the polarization of the material :

$$\boldsymbol{P}_{ind} = \varepsilon_0(\chi^{(1)} \cdot \boldsymbol{E} + \chi^{(2)} \cdot \boldsymbol{E}^2 + \chi^{(3)} \cdot \boldsymbol{E}^3 + \ldots)$$

†Griffiths, D. J., (2012) *Introduction to Electrodynamics*, 4th Edition; Pearson
Boyd, R.W., (2008) *Nonlinear Optics*, 3rd Edition; Elsevier

NON-LINEAR OPTICS

As the light intensity increases, this *linear* model begins to breakdown. Instead, we now use a power series to model the polarization of the material:

$$\boldsymbol{P}_{\boldsymbol{ind}} = \varepsilon_0(\chi^{(1)} \cdot \boldsymbol{E} + \chi^{(2)} \cdot \boldsymbol{E}^2 + \chi^{(3)} \cdot \boldsymbol{E}^3 + \ldots)$$

Considering an electric field of the form:

$$\boldsymbol{E}(t) = E_\omega \cos(\omega t)$$

We obtain for the induced polarizability:

$$\boldsymbol{P}_{\boldsymbol{ind}} = \varepsilon_0(\chi^{(1)} \cdot E_\omega \cos(\omega t) + \chi^{(2)} \cdot E_\omega^2 \cos^2(\omega t) + \chi^{(3)} \cdot E_\omega^3 \cos^3(\omega t) + \ldots)$$

Non-centrosymmetric materials ($\chi^{(3)} = 0$)

$$\cos^2\theta = \frac{1}{2}(1 + \cos\theta)$$

$$\boldsymbol{P}_{\boldsymbol{ind}} = \varepsilon_0(\underbrace{\chi^{(1)} \cdot E_\omega \cos(\omega t)}_{\text{Term oscillating at frequency } \omega \text{ (linear propagation)}} + \underbrace{\frac{1}{2}\chi^{(2)} \cdot E_\omega^2}_{\text{DC non-oscillating term}} + \underbrace{\frac{1}{2}\chi^{(2)} \cdot E_\omega^2 \cos(2\omega t)}_{\text{Term oscillating at frequency } 2\omega \text{ (non-linear generation)}})$$

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NON-LINEAR OPTICS

As the light intensity increases, this *linear* model begins to breakdown. Instead, we now use a power series to model the polarization of the material:

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NON-LINEAR OPTICS

As the light intensity increases, this *linear* model begins to breakdown. Instead, we now use a power series to model the polarization of the material:

$$\boldsymbol{P_{ind}} = \varepsilon_0(\chi^{(1)} \cdot \boldsymbol{E} + \chi^{(2)} \cdot \boldsymbol{E}^2 + \chi^{(3)} \cdot \boldsymbol{E}^3 + \ldots)$$

Considering an electric field of the form:

$$\boldsymbol{E}(t) = E_\omega \cos(\omega t)$$

We obtain for the induced polarizability:

$$\boldsymbol{P_{ind}} = \varepsilon_0(\chi^{(1)} \cdot E_\omega \cos(\omega t) + \chi^{(2)} \cdot E_\omega^2 \cos^2(\omega t) + \chi^{(3)} \cdot E_\omega^3 \cos^3(\omega t) + \ldots)$$

Implies that three fields interact to produce a fourth field. Hence, $\chi^{(3)}$ interactions are four photon processes!

Centrosymmetric materials ($\chi^{(2)} = 0$)

$$\cos^3(\theta) = \frac{1}{4}(3\cos(\theta) + \cos(3\theta))$$

$$\boldsymbol{P_{ind}} = \varepsilon_0(\underbrace{\chi^{(1)} \cdot E_\omega \cos(\omega t)} + \underbrace{\frac{3}{4}\chi^{(3)} \cdot E_\omega^3 \cos(\omega t)} + \underbrace{\frac{1}{4}\chi^{(3)} \cdot E_\omega^3 \cos(3\omega t)})$$

Term oscillating at frequency ω (linear propagation)

Term oscillating at frequency ω (linear generation)

Term oscillating at frequency 3ω (non-linear generation)

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NON-LINEAR OPTICS

- We know that, for the electric displacement, we have for a linear system:

$$\begin{aligned} \boldsymbol{D} &= \varepsilon_0 \cdot \boldsymbol{E} + \boldsymbol{P_{ind}} \\ &= \varepsilon_0 \cdot \boldsymbol{E} + \varepsilon_0 \cdot \chi^{(1)} \cdot \boldsymbol{E} \\ &= \varepsilon_0 \cdot (1 + \chi^{(1)}) \cdot \boldsymbol{E} \\ &= \varepsilon_0 \cdot \varepsilon_r \cdot \boldsymbol{E} \end{aligned}$$

where ε_r is the relative permittivity of the medium.

- We define the refractive index of the medium as:

$$n = \sqrt{\varepsilon_r} = \sqrt{1 + \chi^{(1)}}$$

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NON-LINEAR OPTICS

- Considering a centro-symmetric medium (i.e. $\chi^{(2)} = 0$) the polarization of the material will be:

$$\boldsymbol{P}(\omega) = \varepsilon_0 \cdot \chi^{(1)} \boldsymbol{E}(\omega) + \varepsilon_0 \cdot \chi^{(3)} \boldsymbol{E}^3(\omega)$$
$$= \varepsilon_0 \cdot \boldsymbol{E}(\omega)(\chi^{(1)} + \chi^{(3)} \boldsymbol{E}^2(\omega))$$

- Hence for the electric displacement we will have:

$$\boldsymbol{D}(\omega) = \varepsilon_0 \cdot \boldsymbol{E}(\omega) + \varepsilon_0 \cdot \boldsymbol{E}(\omega)(1 + \chi^{(1)} + \chi^{(3)} \boldsymbol{E}^2(\omega))$$
$$= \varepsilon_0 \cdot \boldsymbol{E}(\omega)(1 + \chi^{(1)} + \chi^{(3)} \boldsymbol{E}^2(\omega))$$

- Noting that $\boldsymbol{E}^2(\omega)$ is simply the intensity $I(\omega)$ of the beam, we can express the relative permittivity of the medium as:

$$\varepsilon_r = (1 + \chi^{(1)} + \chi^{(3)} I(\omega))$$

- This yields for the refractive index of the medium:

$$n = \sqrt{\varepsilon_r} = \sqrt{(1 + \chi^{(1)} + \chi^{(3)} I(\omega))}$$

NON-LINEAR OPTICS

- This yields for the refractive index of the medium:

$$n = \sqrt{\varepsilon_r} = \sqrt{\underbrace{(1+\chi^{(1)}}_{n_0} + \chi^{(3)} I(\omega))}$$

- Which we can approximate as:

$$n = \sqrt{\varepsilon_r} \approx n_0 + \frac{1}{2n_0}\chi^{(3)} I(\omega)$$

- We set the *non-linear refractive index* of the material as $\mathrm{n}_2 = \frac{1}{2n_0}\chi^{(3)}$, allowing us to express the, intensity dependent, refractive index as:

$$\boxed{n(I) = n_0 + n_2 I(\omega)}$$

NON-LINEAR OPTICS

TABLE 4.1.2 Third-order nonlinear optical coefficients of various materials [a]

Material	n_0	$\chi^{(3)}$ (m^2/V^2)	n_2 (cm^2/W)	Comments and References [b]
Crystals				
Al_2O_3	1.8	3.1×10^{-22}	2.9×10^{-16}	1
CdS	2.34	9.8×10^{-20}	5.1×10^{-14}	1,1.06 μm
Diamond	2.42	2.5×10^{-21}	1.3×10^{-15}	1
GaAs	3.47	1.4×10^{-18}	3.3×10^{-13}	1,1.06 μm
Ge	4.0	5.6×10^{-19}	9.9×10^{-14}	2, THG $\lvert\chi^{(3)}\rvert$
LiF	1.4	6.2×10^{-23}	9.0×10^{-17}	1
Si	3.4	2.8×10^{-18}	2.7×10^{-14}	2, THG $\lvert\chi^{(3)}\rvert$
TiO_2	2.48	2.1×10^{-20}	9.4×10^{-15}	1
ZnSe	2.7	6.2×10^{-20}	3.0×10^{-14}	1,1.06 μm
Glasses				
Fused silica	1.47	2.5×10^{-22}	3.2×10^{-16}	1
As_2S_3 glass	2.4	4.1×10^{-19}	2.0×10^{-13}	3
BK-7	1.52	2.8×10^{-22}	3.4×10^{-16}	1
BSC	1.51	5.0×10^{-22}	6.4×10^{-16}	1
Pb Bi gallate	2.3	2.2×10^{-20}	1.3×10^{-14}	4
SF-55	1.73	2.1×10^{-21}	2.0×10^{-15}	1
SF-59	1.953	4.3×10^{-21}	3.3×10^{-15}	1
Nanoparticles				
CdSSe in glass	1.5	1.4×10^{-20}	1.8×10^{-14}	3, nonres.
CS 3-68 glass	1.5	1.8×10^{-16}	2.3×10^{-10}	3, res.
Gold in glass	1.5	2.1×10^{-16}	2.6×10^{-10}	3, res.
Polymers				
Polydiacetylenes				
PTS		8.4×10^{-18}	3.0×10^{-12}	5, nonres.
PTS		-5.6×10^{-16}	-2.0×10^{-10}	6, res.
9BCMU			2.7×10^{-18}	7, $\lvert n_2\rvert$, res.
4BCMU	1.56	-1.3×10^{-19}	-1.5×10^{-13}	8, nonres, $\beta =$

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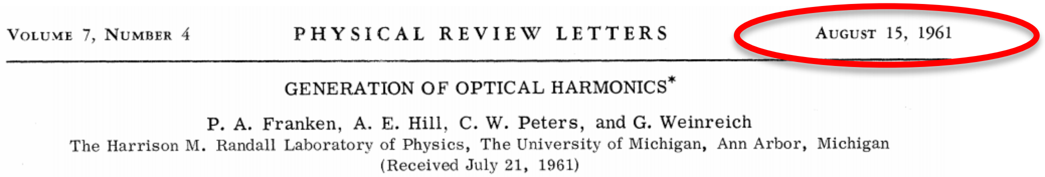
NON-LINEAR OPTICS

Maria Goeppert Mayer predicts two photon absorption as part of her PhD – 1931

Theodore Maiman and the first laser (Ruby) – May 1960

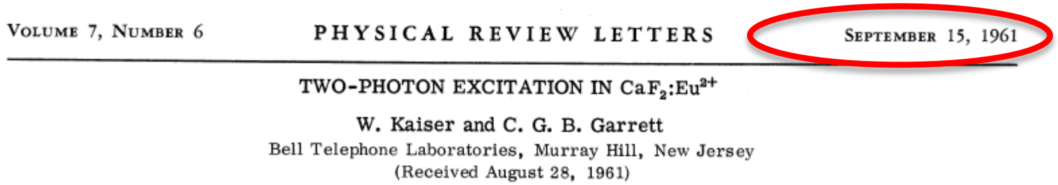


VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

Second Harmonic Generation – 1961

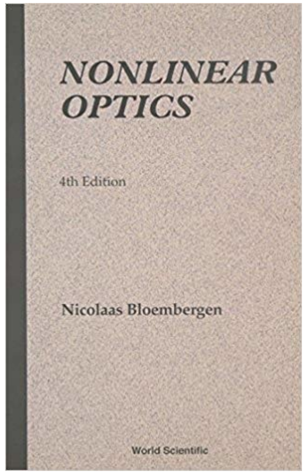


VOLUME 7, NUMBER 6 PHYSICAL REVIEW LETTERS SEPTEMBER 15, 1961

TWO-PHOTON EXCITATION IN $CaF_2:Eu^{2+}$

W. Kaiser and C. G. B. Garrett
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received August 28, 1961)

Two photon absorption – 1961



Nicolaas Bloembergen, Nobel Laurate in Physics 1981, wrote *NonLinear Optics* in 1964

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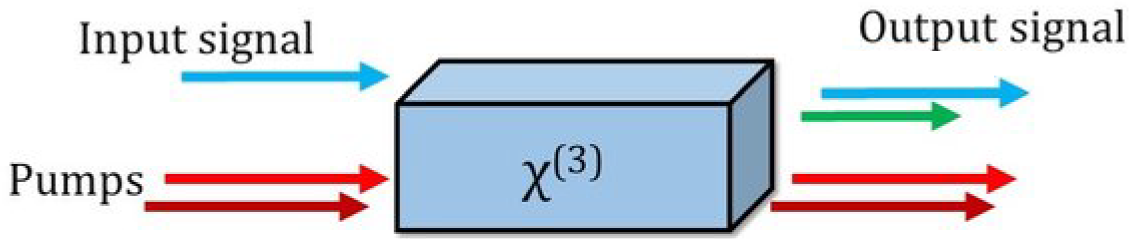
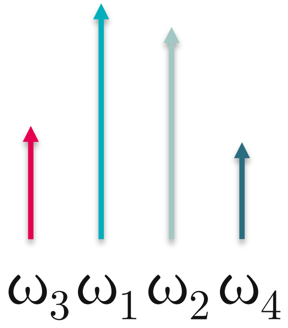
FOUR-WAVE MIXING





<u>Conservation of energy</u>:

$$\omega_4 = \pm\omega_1 \pm \omega_2 \pm \omega_3$$

<u>Conservation of momentum</u>:

$$\boldsymbol{k}_4 = \pm\boldsymbol{k}_1 \pm \boldsymbol{k}_2 \pm \boldsymbol{k}_3$$

Phase-matching (Δk=0)

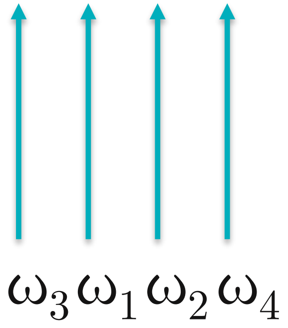
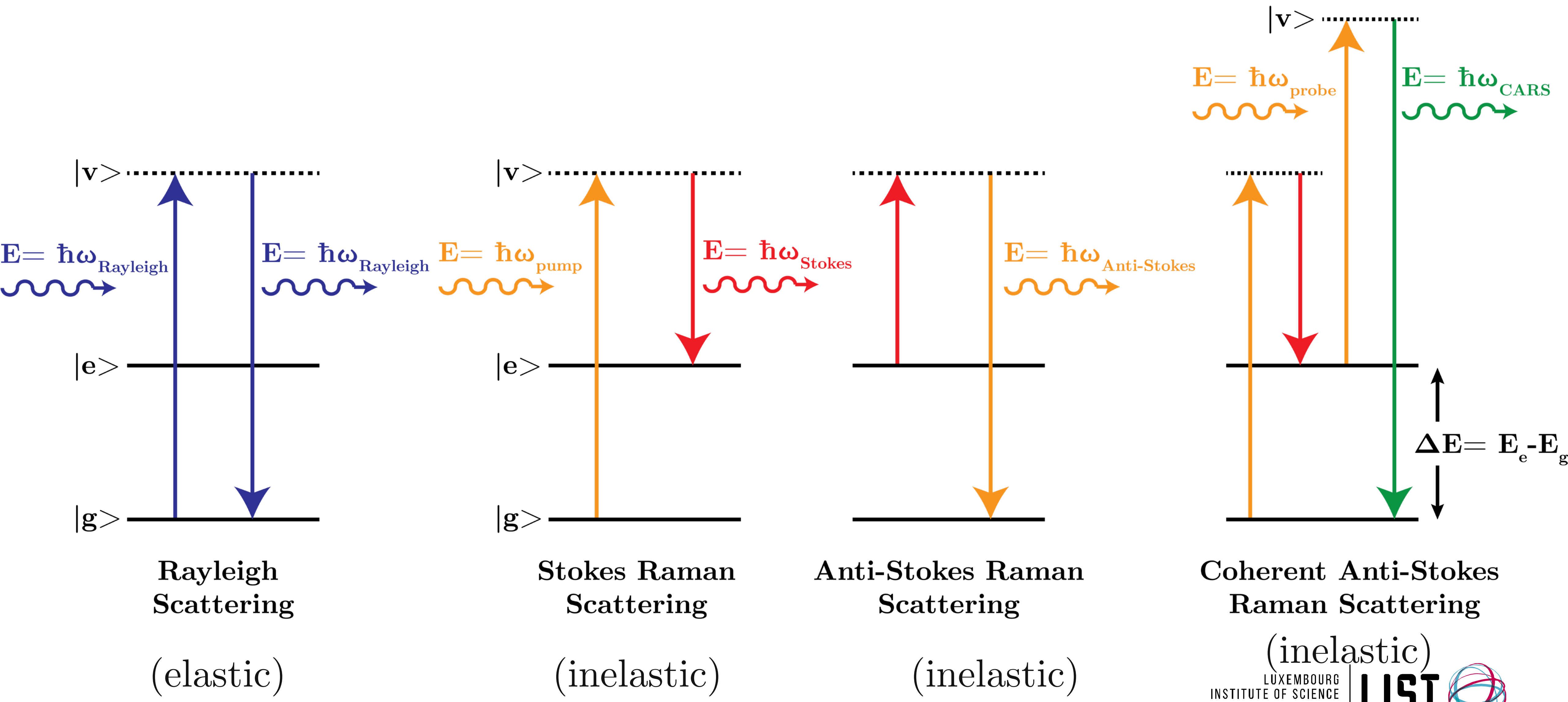


Image from Bryn A. Bell, Kai Wang, Alexander S. Solntsev, Dragomir N. Neshev, Andrey A. Sukhorukov, and Benjamin J. Eggleton, "Spectral photonic lattices with complex long-range coupling," Optica 4, 1433-1436 (2017)

COHERENT ANTI-STOKES RAMAN SCATTERING



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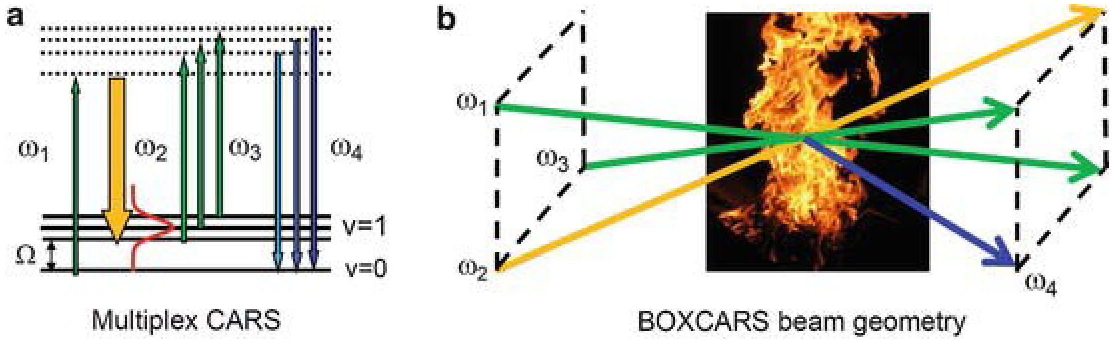
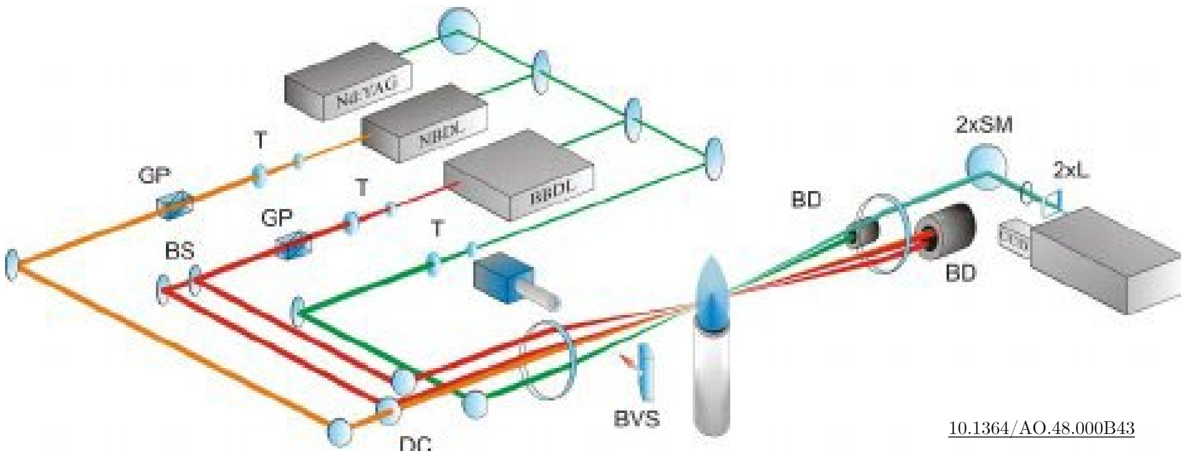
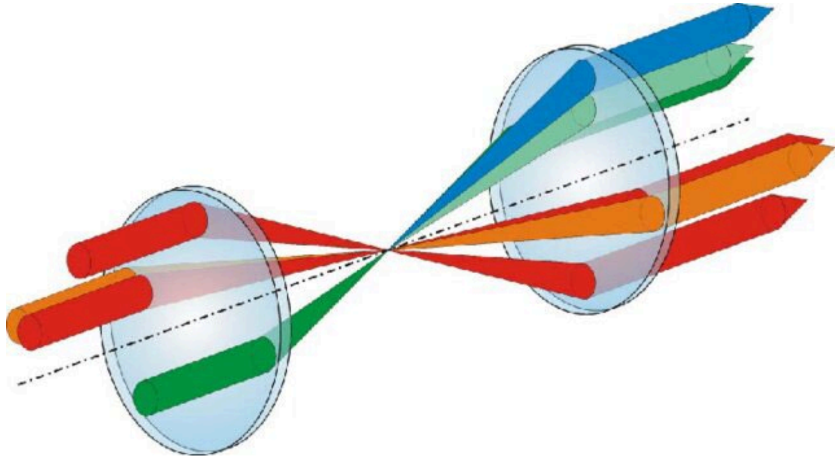
COHERENT ANTI-STOKES RAMAN SCATTERING



https://link.springer.com/referenceworkentry/10.1007/978-3-642-16712-6_136





10.1364/AO.48.000B43

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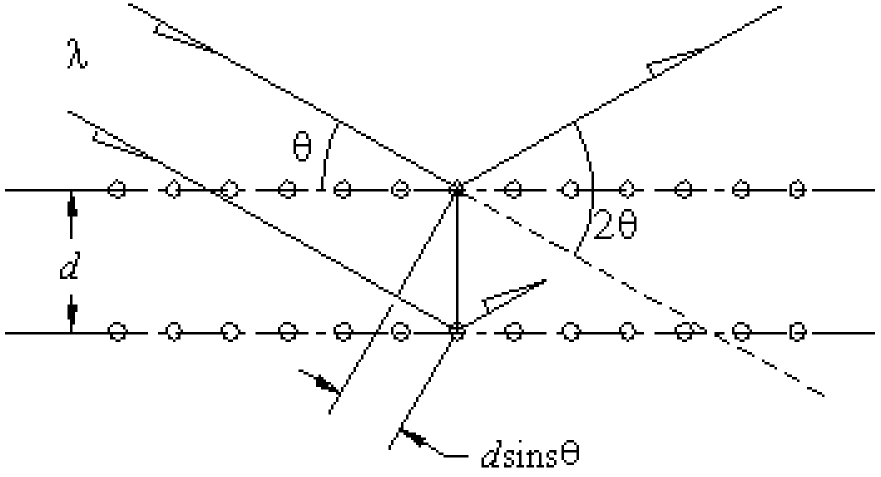
SOME PHYSICS…

BRAGG SCATTERING



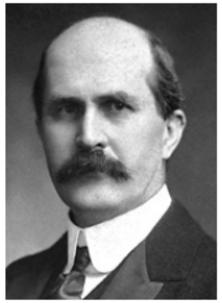
evident that the peaks $A_1 B_1 C_1$, $A_2 B_2 C_2$ are analogous to spectra of the first and second orders, because of the absence of intervening sets of peaks. The value of n in the equation

$$n\lambda = 2d \sin\theta$$

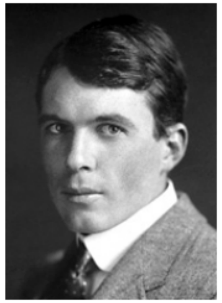
seems clear. The difficulty of assigning a definite wave-length to the rays arises when we attempt to determine the value of d, the distance of plane from plane.

W. H. Bragg, W. L. Bragg, The Reflection of X-rays by Crystals, Proc. R. Soc. Lond. A 1913 88 428-438; DOI: 10.1098/rspa.1913.0040. Published 1 July 1913

The Nobel Prize in Physics 1915



Sir William Henry Bragg
Prize share: 1/2



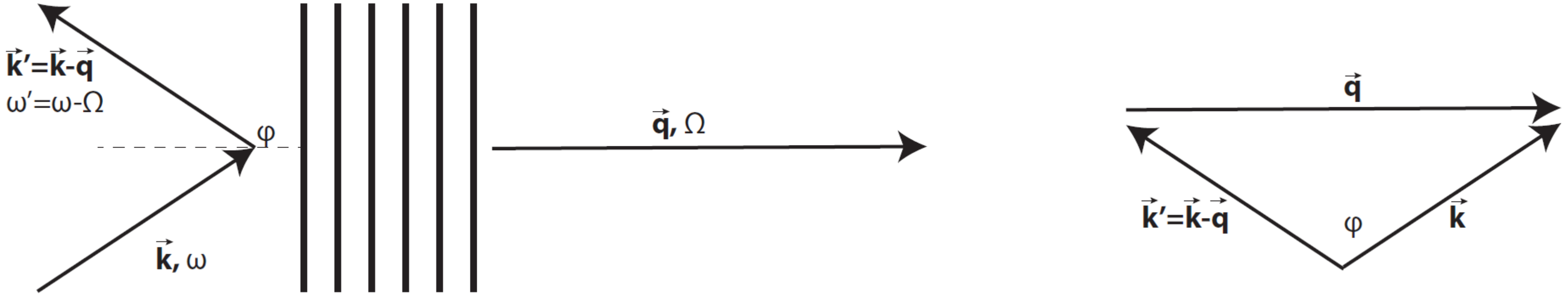
William Lawrence Bragg
Prize share: 1/2

The Nobel Prize in Physics 1915 was awarded jointly to Sir William Henry Bragg and William Lawrence Bragg *"for their services in the analysis of crystal structure by means of X-rays"*

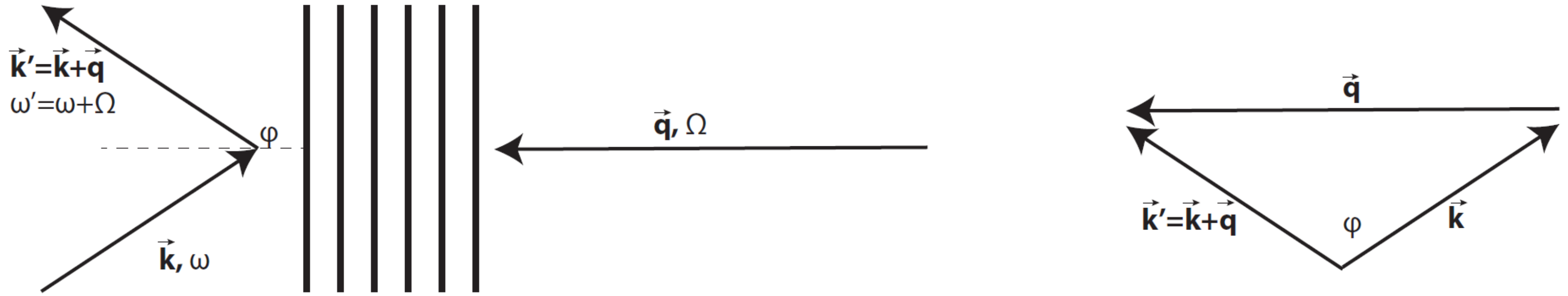
*x-ray crystallography was also used to determine the double helix structure of the DNA molecule (1962 Nobel Prize in Physiology), haemoglobin (1962 Nobel Prize in Chemistry) and other proteins

BRILLOUIN SCATTERING

Stokes Brillouin peak:

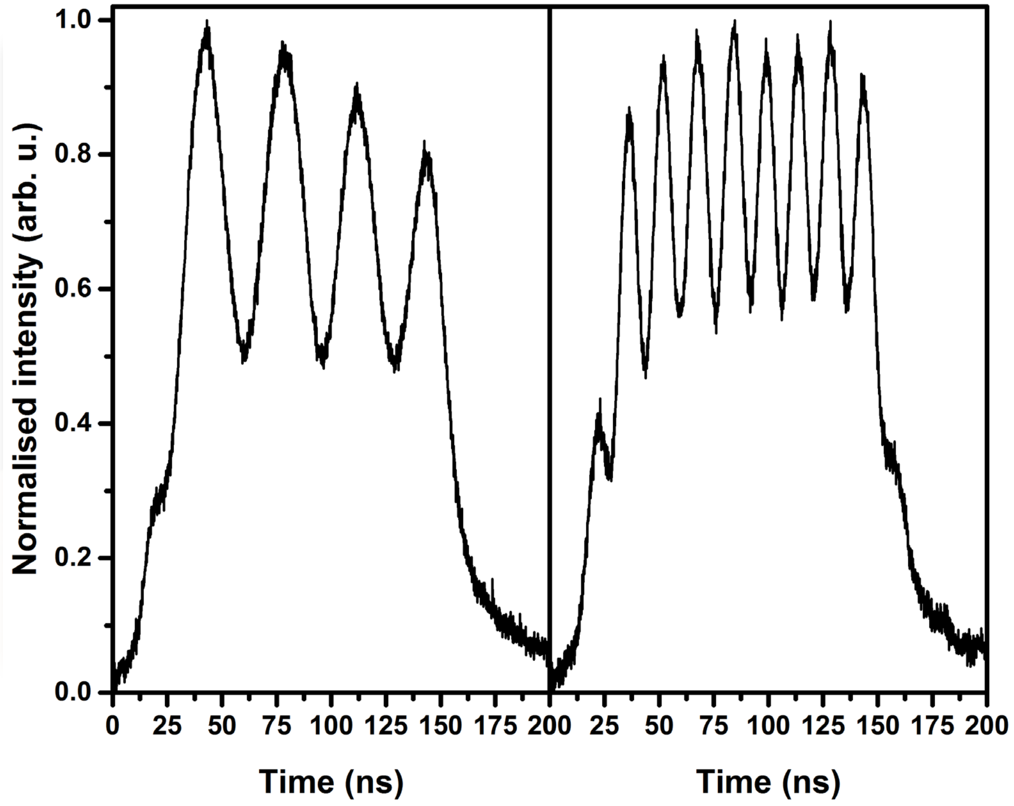


Anti-Stokes Brillouin peak:



L. Brillouin, Annales de Physique (Paris) 17 (1922)

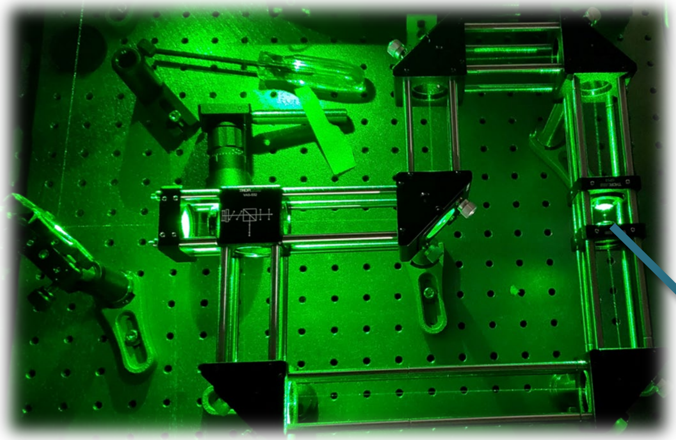
ELECTROSTRICTION

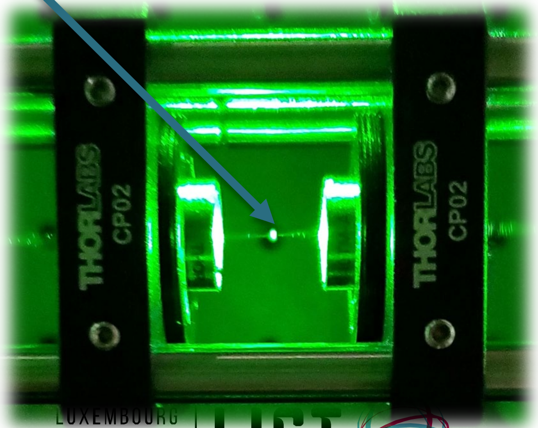


Force on a particle in an electric field:

$$F = -\nabla U = \frac{1}{2}\epsilon_0 \alpha_{eff} \nabla(E^2)$$

$$\alpha_{eff} = [(\alpha_{\parallel} - \alpha_{\perp})cos^2\theta + \alpha_{\perp}]$$



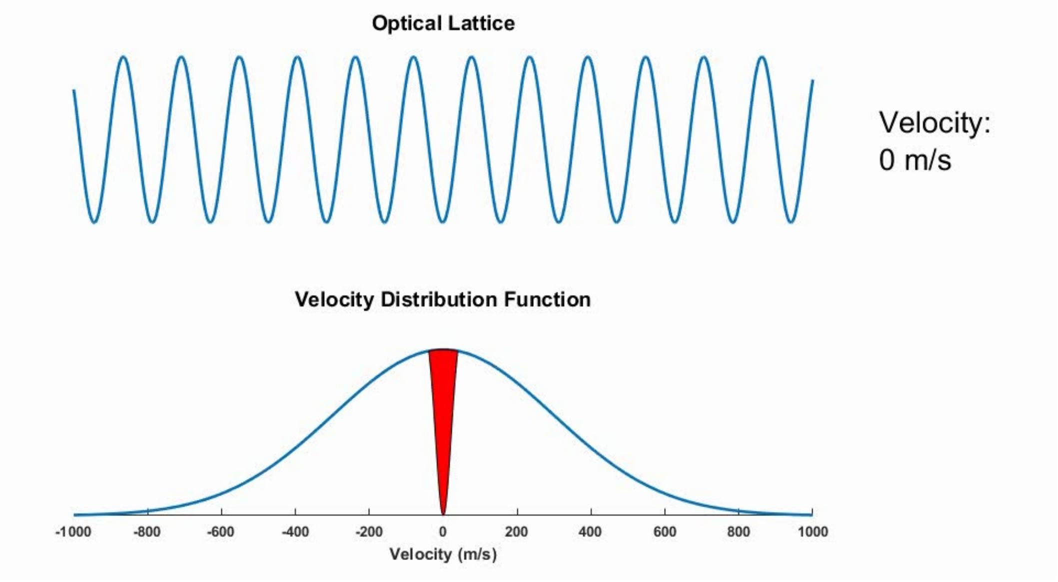


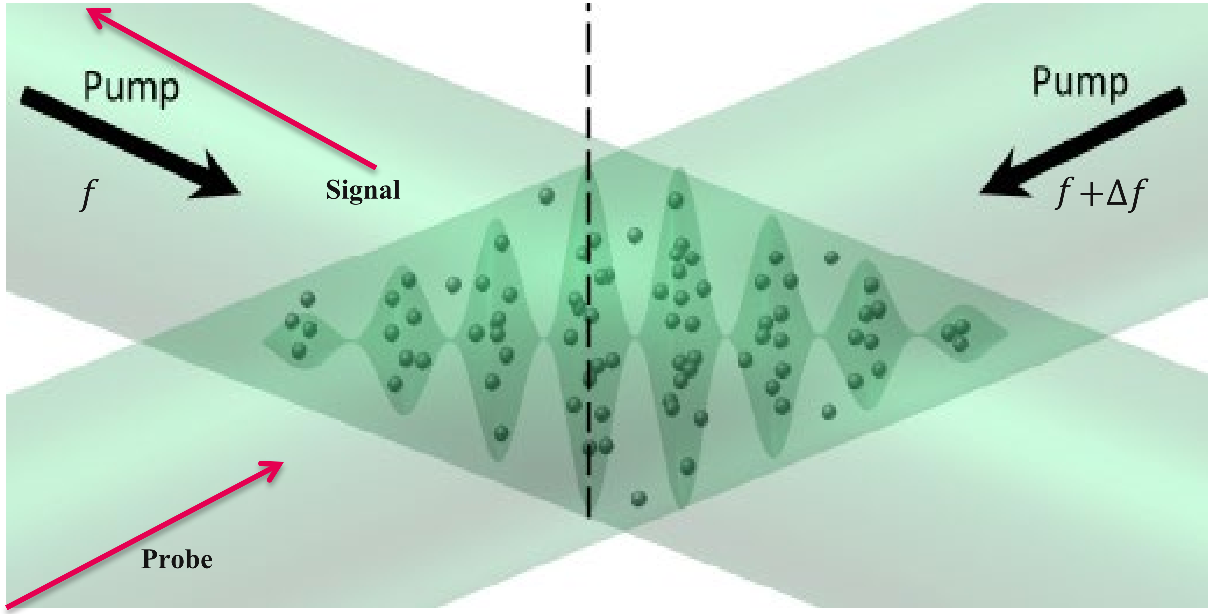
"The tendency of polarizable molecules to move towards regions of high optical electric field intensities is referred to as *electrostriction*."

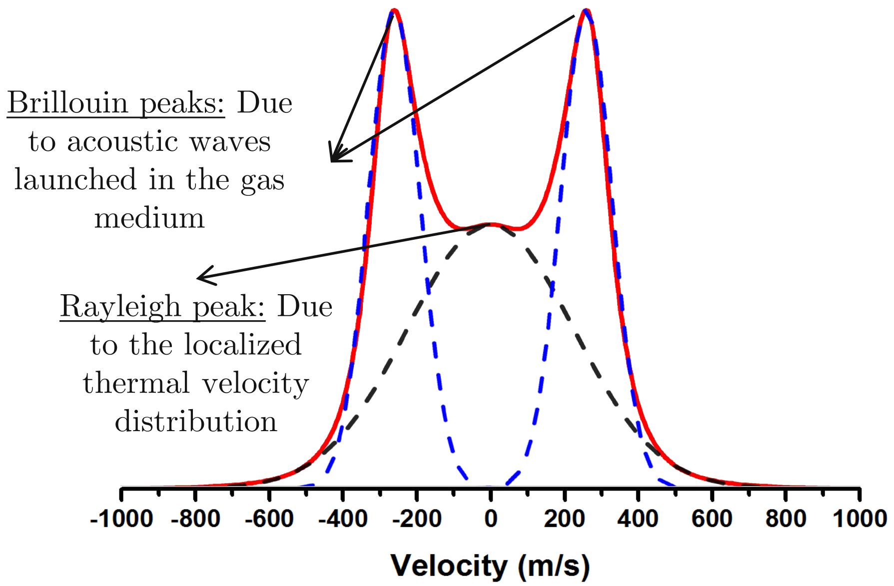
R.W. Boyd, *Nonlinear Optics*. Elsevier, London, 3rd edition, 2008.

Animation taken from http://www.fhi-berlin.mpg.de/mp/meerakker/uploads/Research/animation.gif (accessed 4/22/2018)

COHERENT RAYLEIGH-BRILLOUIN SCATTERING







Measurable quantities

Temperature
Density
Speed of sound
Shear & Bulk viscosity
Polarizability

CRBS:

- ✓ Highly localized
- ✓ Laser beam signal output
- ✓ Much lower resolvable densities

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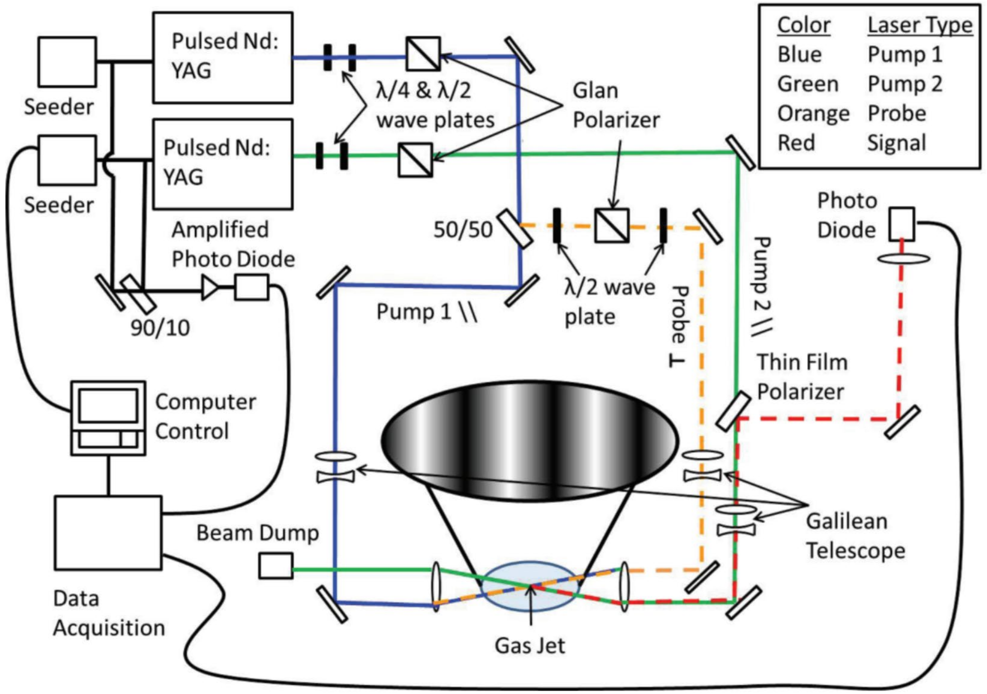
COHERENT RAYLEIGH-BRILLOUIN SCATTERING



VOLUME 85, NUMBER 6 PHYSICAL REVIEW LETTERS 7 AUGUST 2000

Coherent Rayleigh Scattering

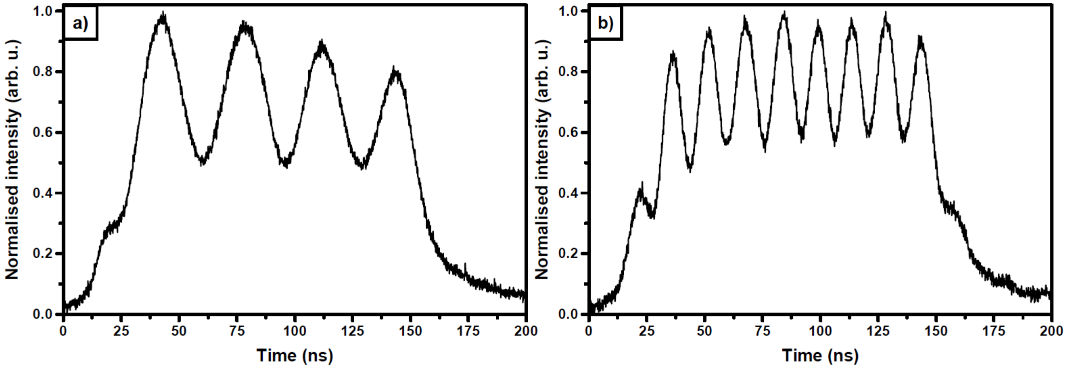
Jay H. Grinstead[1] and Peter F. Barker[2]
[1]Optical Technology Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899
[2]Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544
(Received 21 January 2000)

VOLUME 89, NUMBER 18 PHYSICAL REVIEW LETTERS 28 OCTOBER 2002

Coherent Rayleigh-Brillouin Scattering

Xingguo Pan, Mikhail N. Shneider, and Richard B. Miles
Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey 08544
(Received 26 April 2002; published 10 October 2002)

Constant velocity lattices



Coherent Brillouin scattering

A. Gerakis,[1] M. N. Shneider,[2] and P. F. Barker[1]
[1]Department of Physics and Astronomy, University College London, WC1E 6BT, United Kingdom
[2]Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, New Jersey,08544, USA

PHYSICAL REVIEW A **87**, 033825 (2013)

Narrowband coherent Rayleigh-Brillouin scattering from gases confined by a high-intensity optical lattice

Barry M. Cornella and Sergey F. Gimelshein
ERC Inc., Edwards AFB, California 93524, USA

Taylor C. Lilly
Department of Mechanical and Aerospace Engineering, University of Colorado Colorado Springs, Colorado Springs, Colorado 80918, USA

Andrew D. Ketsdever
Air Force Research Laboratory, Edwards AFB, California 93524, USA
(Received 12 January 2013; published 22 March 2013)

COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Single Shot

Chirped lattice

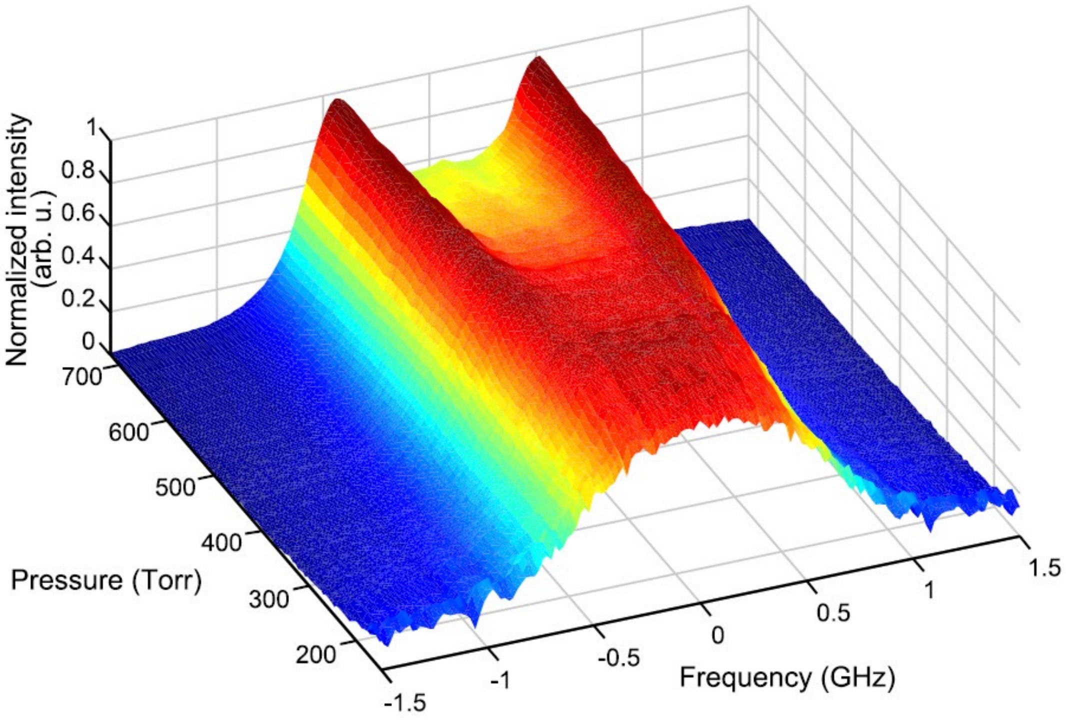
CRBS:

- ✓ Highly localized
- ✓ Laser beam signal output
- ✓ Much lower resolvable densities
- ✓ Single Shot

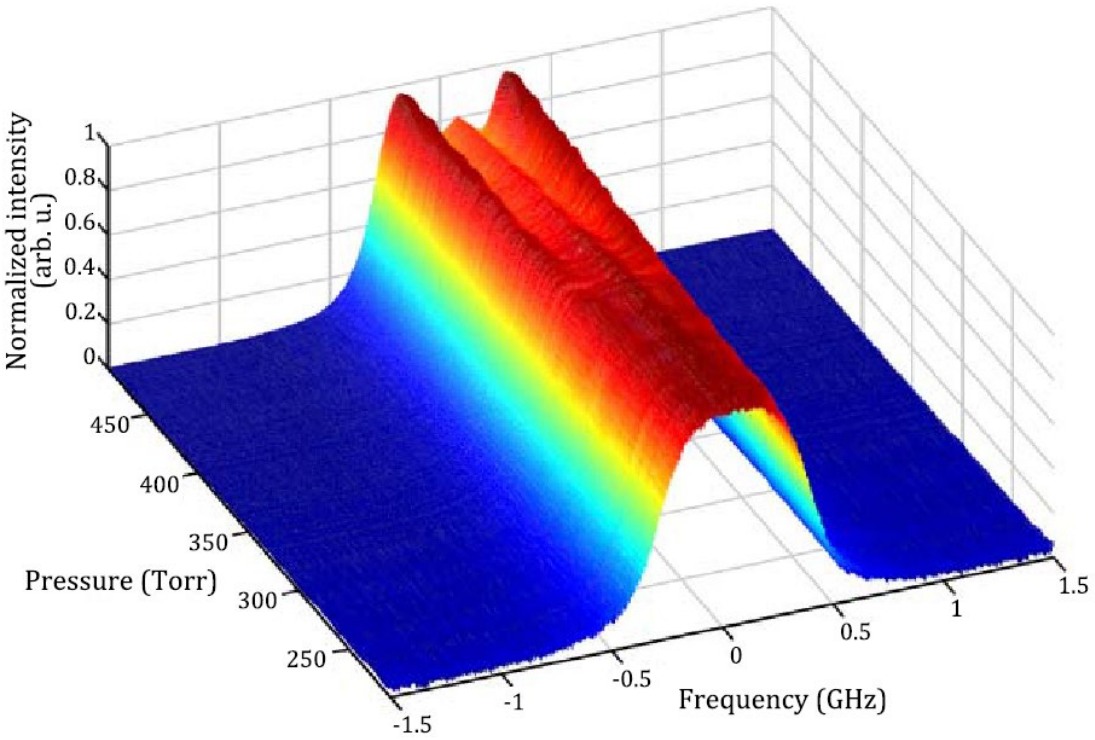
Gerakis, A., Shneider, M.N. and Barker, P.F., 2013. Single-shot coherent Rayleigh–Brillouin scattering using a chirped optical lattice. Optics letters, 38(21), pp.4449-4452.

COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Single Shot



CO_2



Xenon

Gerakis, A., Shneider, M.N. and Barker, P.F., 2013. Single-shot coherent Rayleigh–Brillouin scattering using a chirped optical lattice. Optics letters, 38(21), pp.4449-4452.

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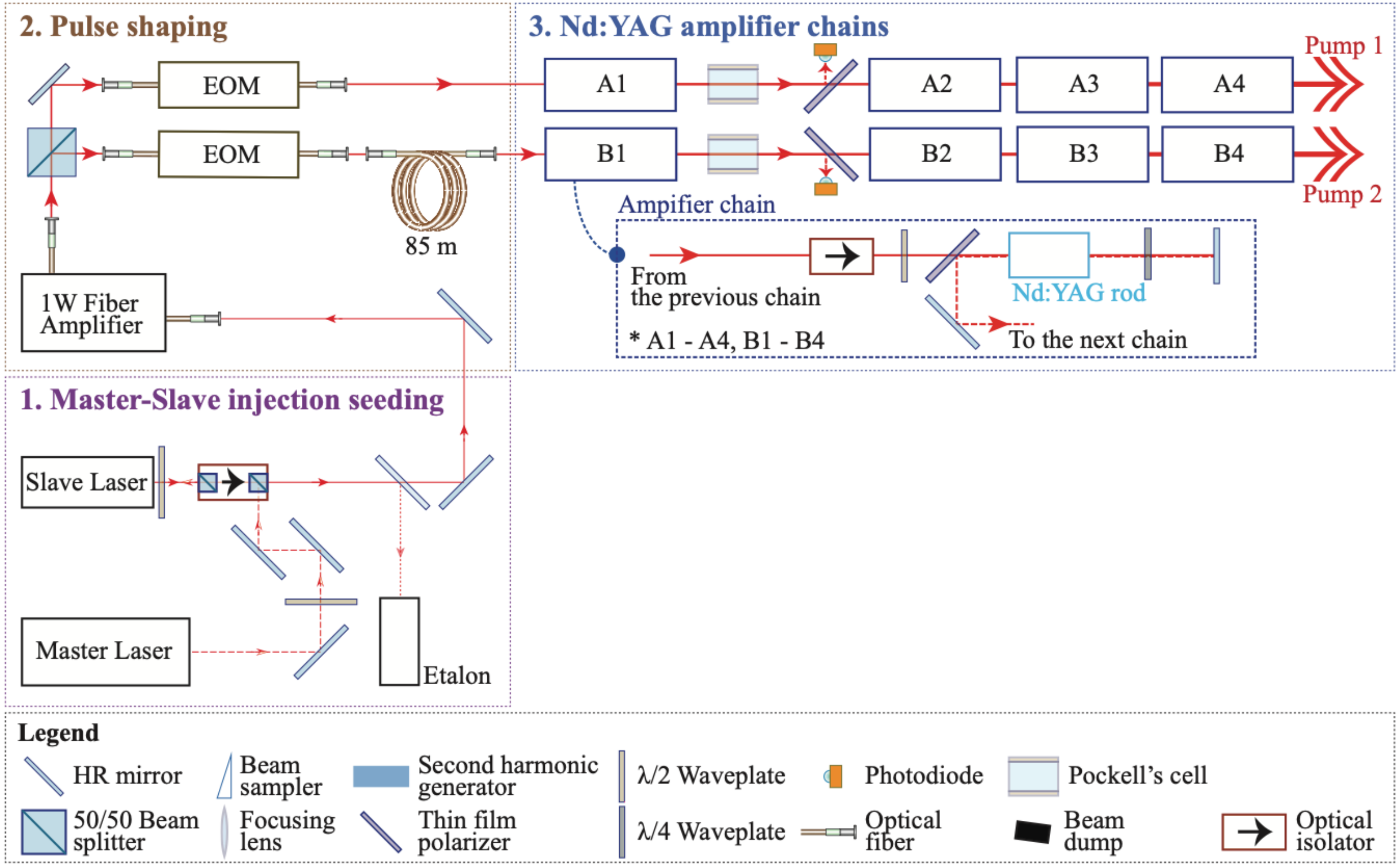
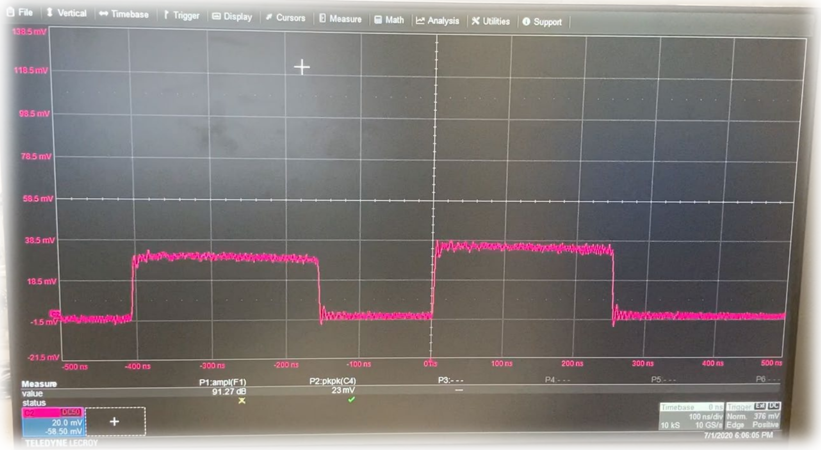
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

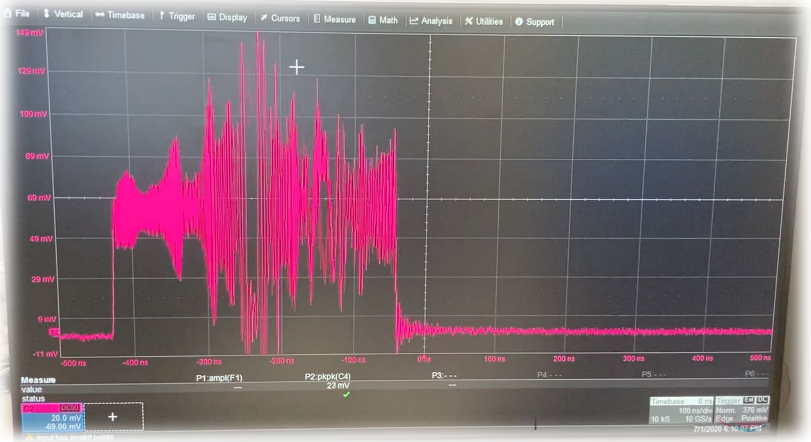
Experimental Setup

COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Experimental Setup







Research Article Vol. 0, No. 0/ 00 00 0000 / Optics Express 1

Optics EXPRESS

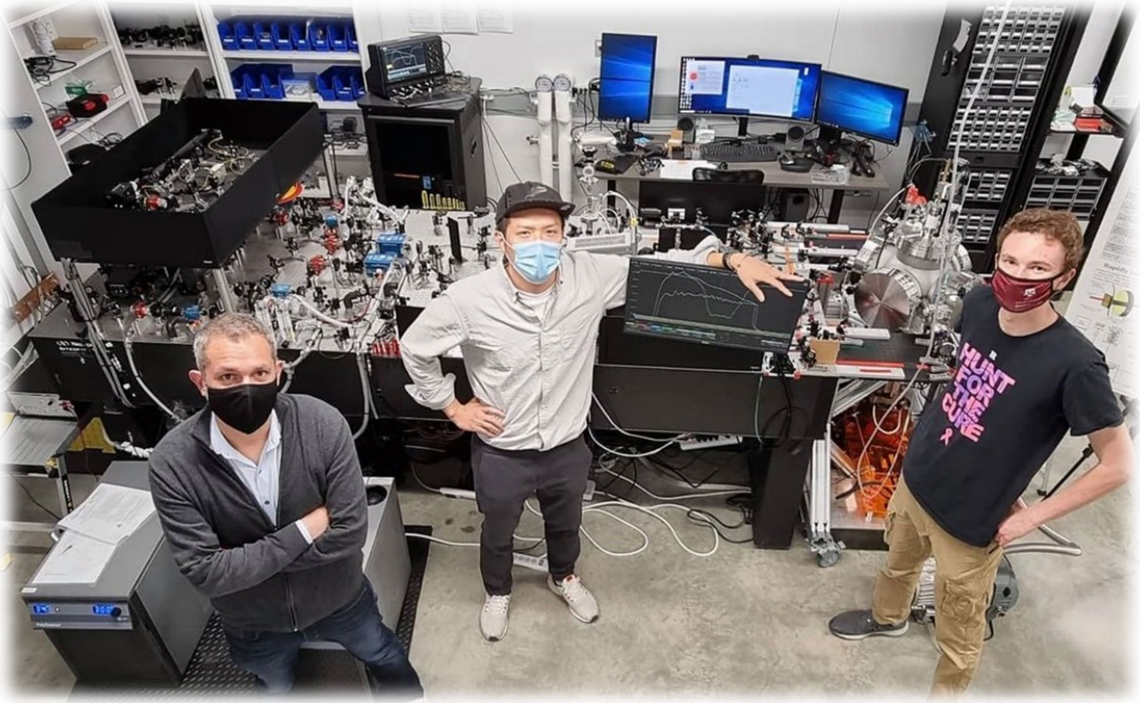
Dual color, frequency, pulse duration and shape agile laser system for particle spectroscopy and manipulation

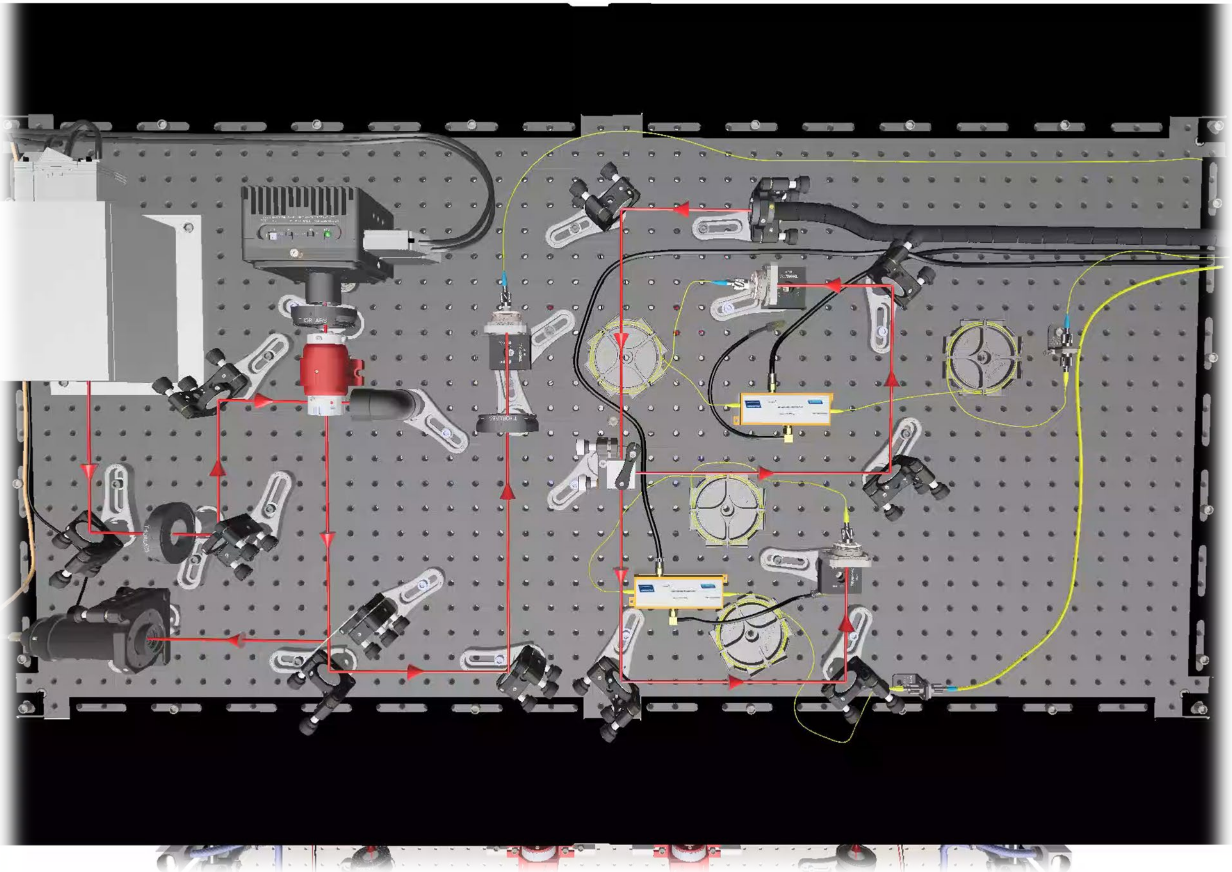
JUNHWI BAK,[1] ROBERT RANDOLPH,[1] AND ALEXANDROS GERAKIS[1,2,*]

COHERENT RAYLEIGH-BRILLOUIN SCATTERING
Experimental Setup

Experimental parameters

- >450 mJ/pulse/arm
- Up to ~10 GHz chirp rate
- Variable pulse durations: 10-1000ns
- Rep. Rate: 10-30 Hz





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COHERENT RAYLEIGH-BRILLOUIN SCATTERING

In-situ measurement of nanoparticles (<10 nm) in arc discharge

Arc method :

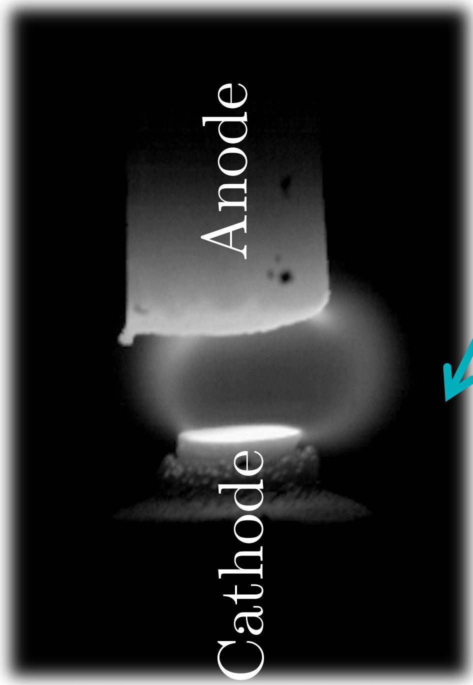
- Simple to implement
- High nanomaterial yield
- Variety of synthesized nanostructures

Environmental conditions :

Helium pressure	~500 Torr
Electrodes	Carbon, <1 cm in diameter
Power	few kW
Plasma density	$10^{14} – 10^{16}$ cm^{-3}
Temperature	~1 eV

Plasma role in nanostructure synthesis?

- What are plasma properties?
- How feedstock material is formed?
- What growth conditions are realized in the arc?



Imaging through C2 filter

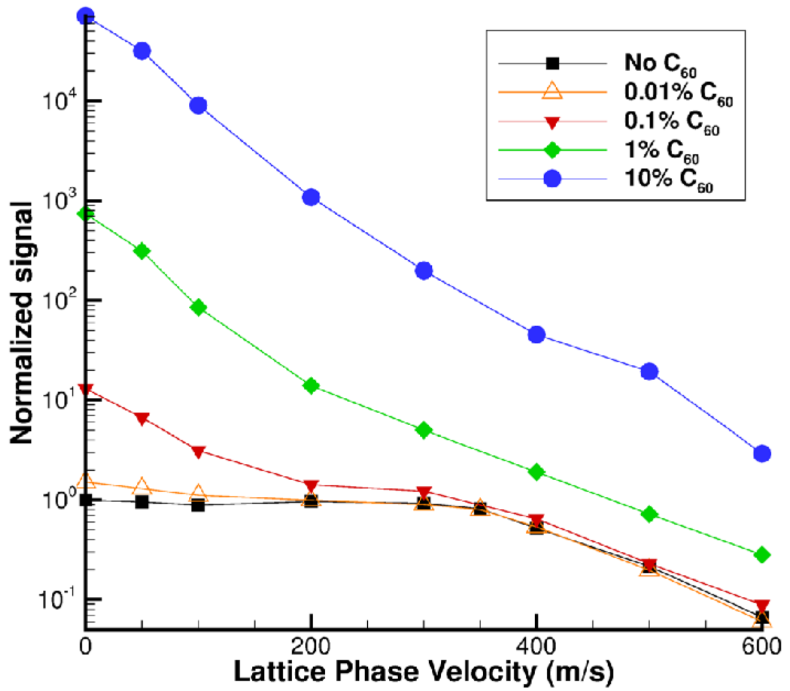
Particles are formed here

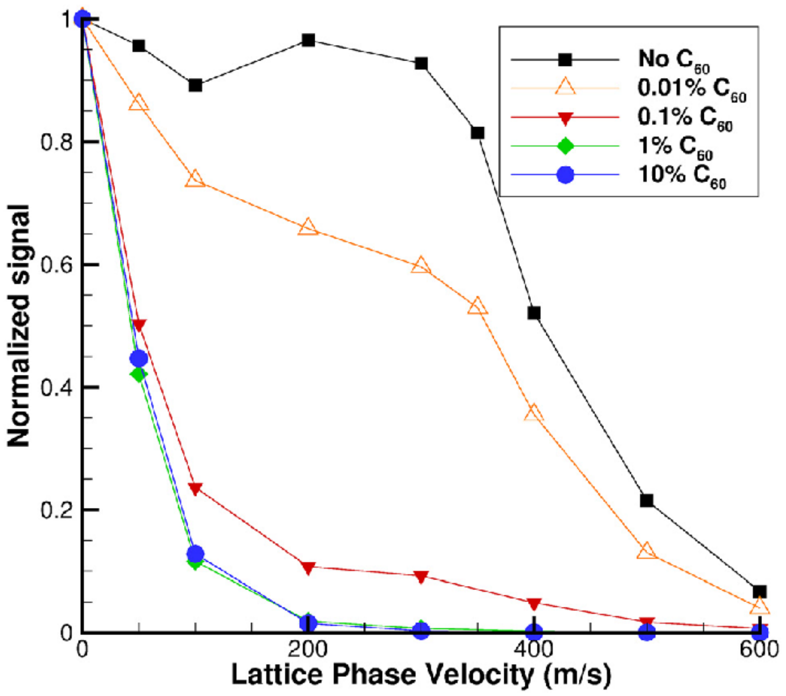
Goal: create prescribed nanoparticles

- Model arc synthesis: plasma modeling and growth modeling
- <u>*In situ*</u> diagnostics to validate model
  - No optical way to measure nanoparticles from <u>initial nucleation to ~10nm</u>... unless we use <u>coherent Rayleigh-Brillouin scattering</u>
- Iterate

COHERENT RAYLEIGH-BRILLOUIN SCATTERING

In-situ measurement of nanoparticles (<10 nm) in arc discharge





APPLIED PHYSICS LETTERS **102**, 173109 (2013)

Application of coherent Rayleigh-Brillouin scattering for *in situ* nanoparticle and large molecule detection

M. N. Shneider[1,a)] and S. F. Gimelshein[2]
[1]*Princeton University, D-429 Engineering Quad D-Wing, Princeton, New Jersey 08544, USA*
[2]*University of Southern California, UPC, RRB 104, Los Angeles, California 90089-1191, USA*

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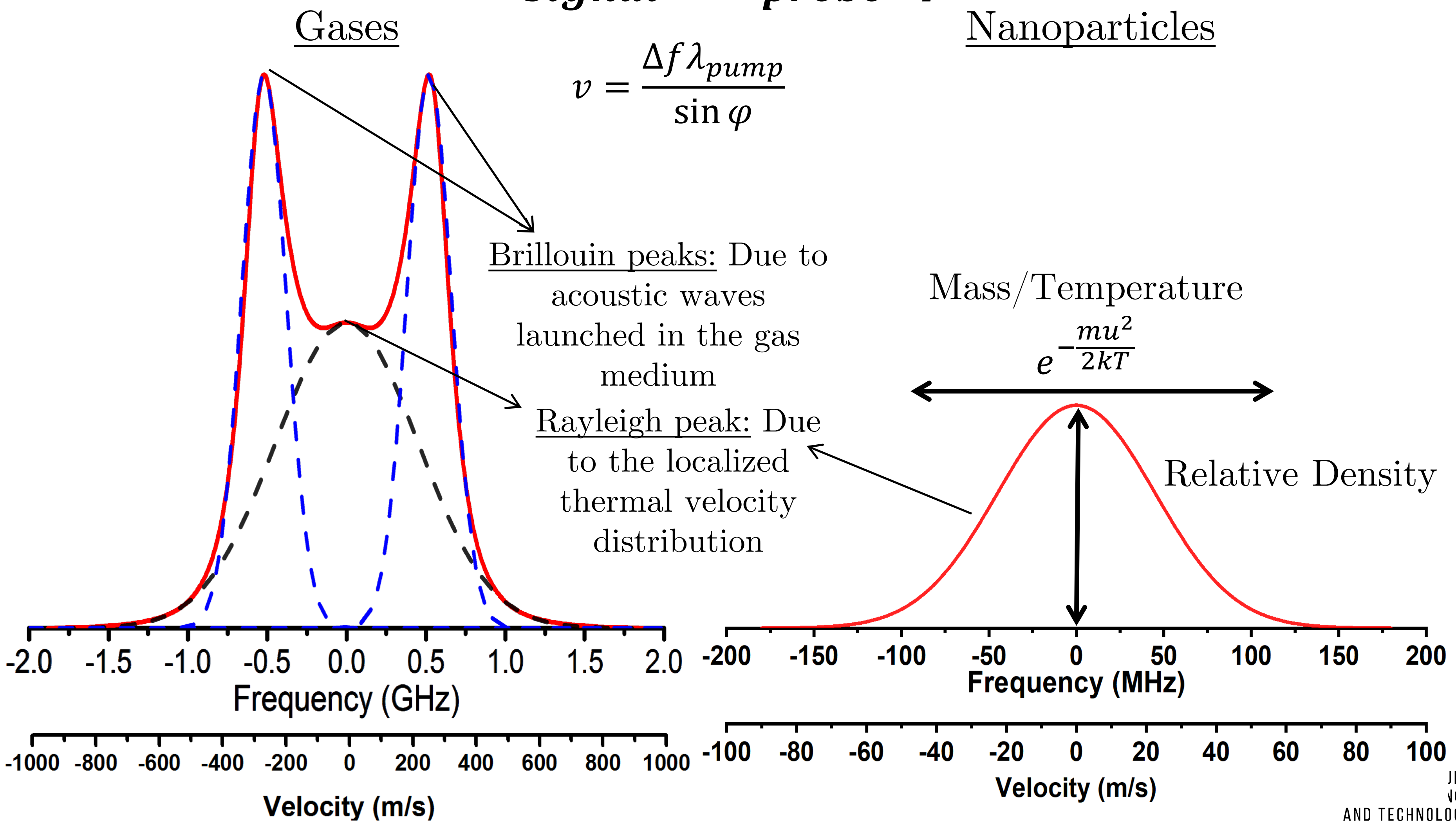
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

In-situ measurement of nanoparticles (<10 nm) in arc discharge

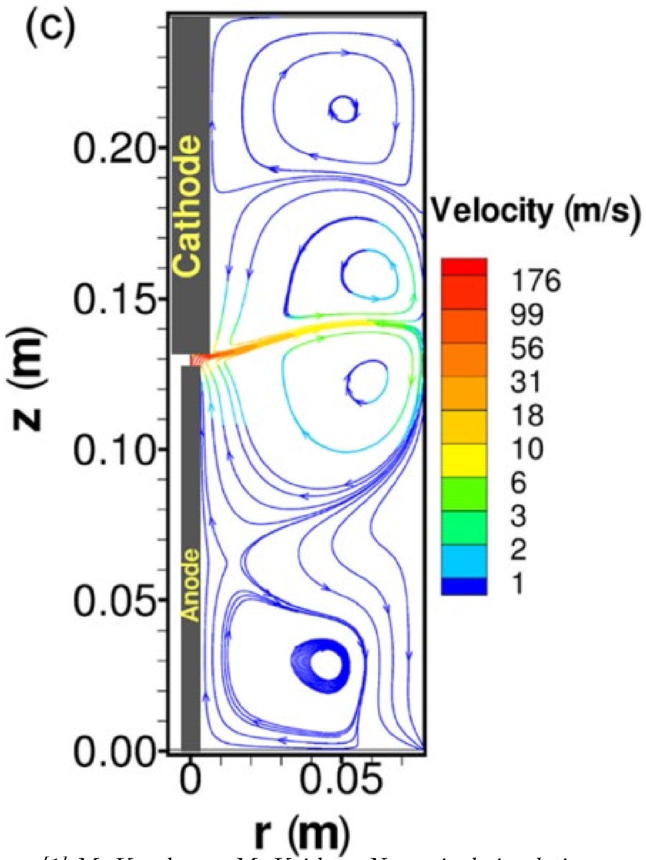
$$I_{signal} \propto I_{probe} \delta\rho^2 L$$

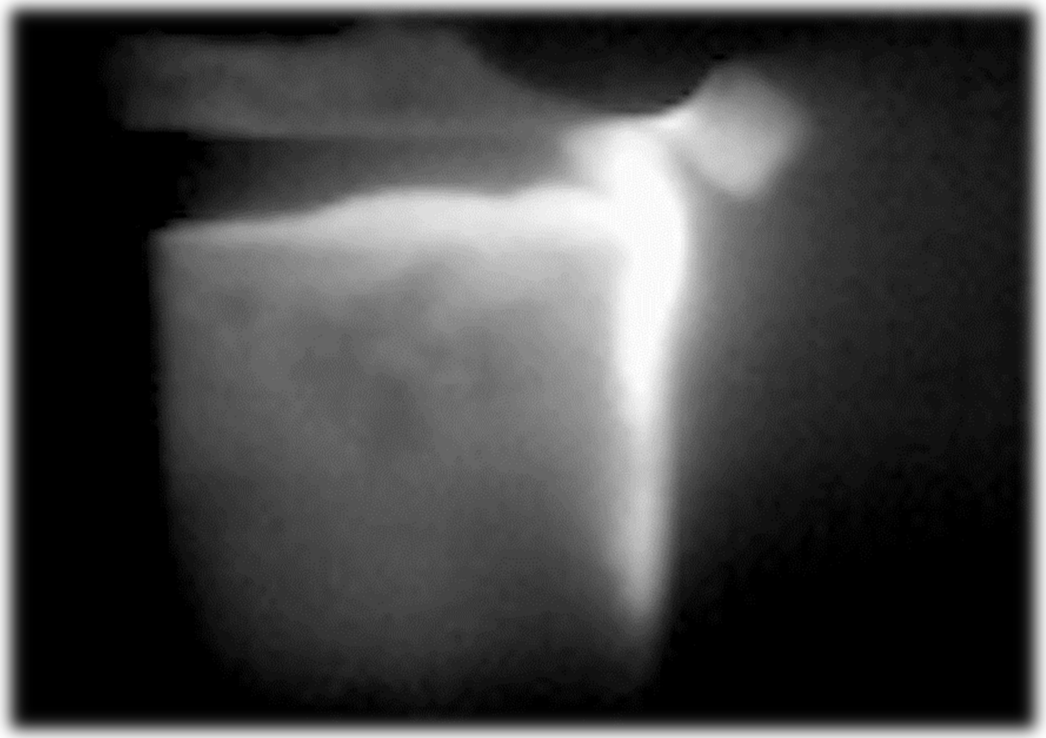


COHERENT RAYLEIGH-BRILLOUIN SCATTERING

In-situ measurement of nanoparticles (<10 nm) in arc discharge

- The arc run time is ~2 minutes
- Nanoparticles have got a residence time of ~1 ms [1]
- Residence time of nanopartilces affects growth

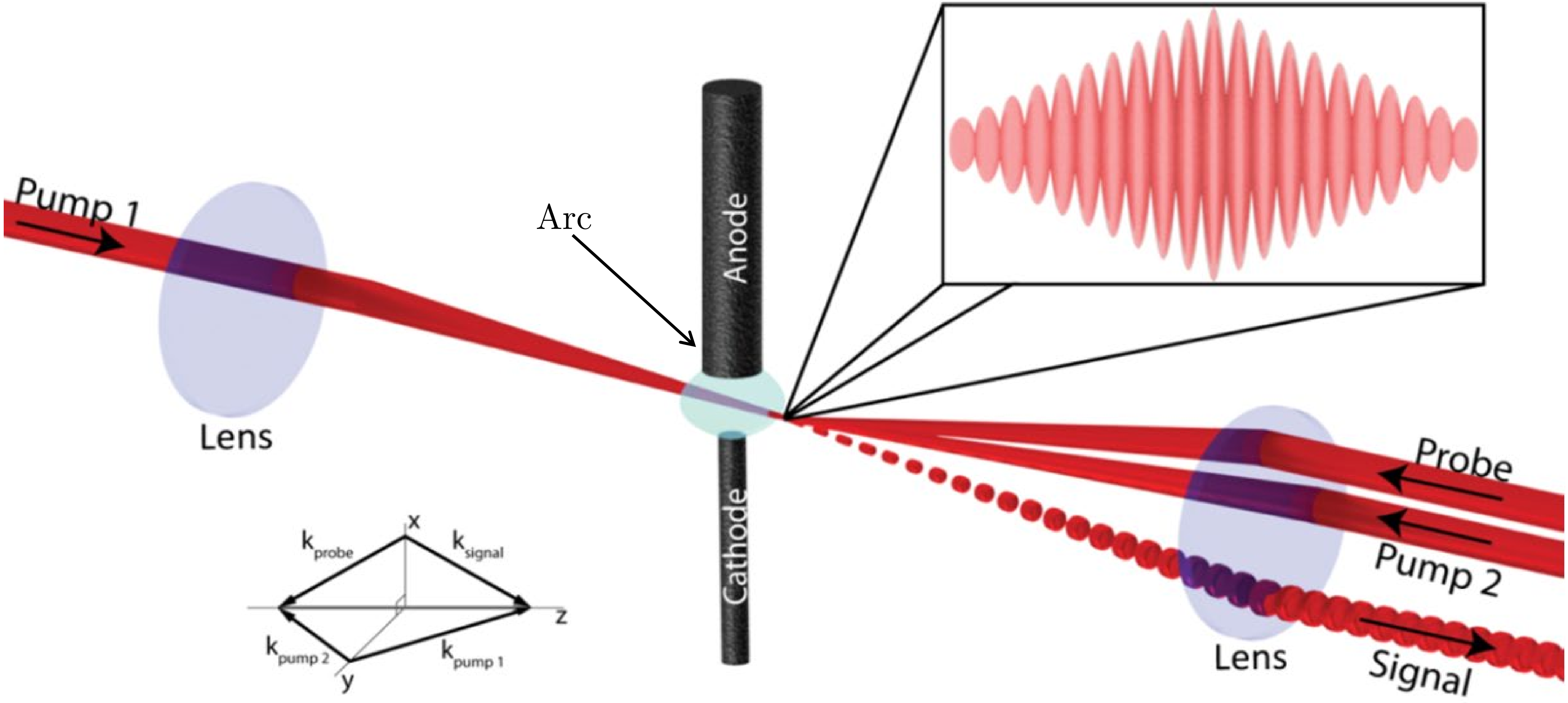




[1] M. Kundrapu, M. Keidar, Numerical simulation of carbon arc discharge for nanoparticle synthesis, Physics of Plasma, Vol. 19, No. 073510, 2012

COHERENT RAYLEIGH-BRILLOUIN SCATTERING

In-situ measurement of nanoparticles (<10 nm) in arc discharge



A. Gerakis, Y-W. Yeh, M. N. Shneider, J. M. Mitrani, B. C. Stratton, and Y. Raitses, "Four-Wave-Mixing Approach to *In Situ* Detection of Nanoparticles", Phys. Rev. Applied 9, 014031 (2018)

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COHERENT RAYLEIGH-BRILLOUIN SCATTERING

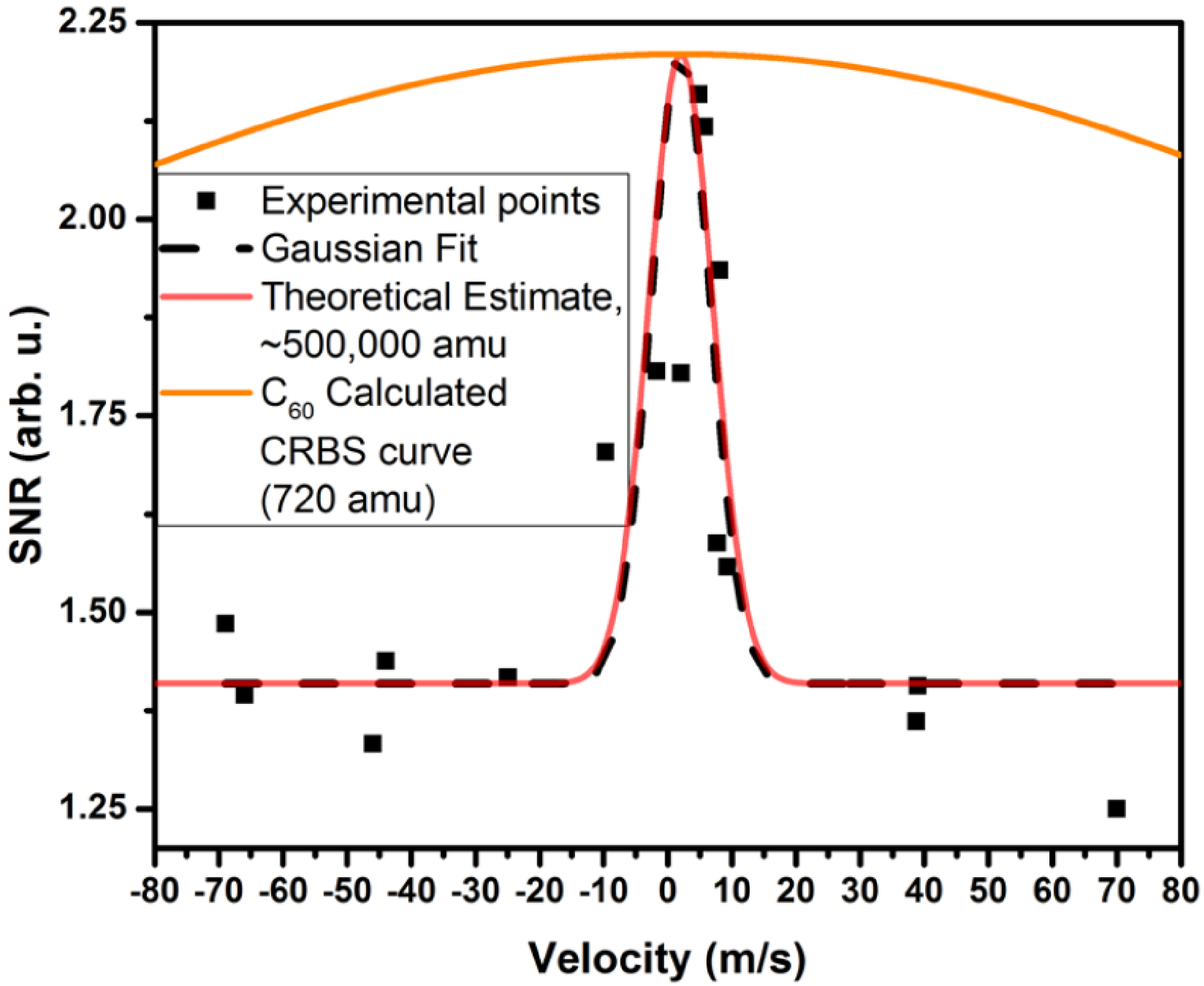
In-situ measurement of nanoparticles (<10 nm) in arc discharge

C_{60} FWHM (@1500 K): 309.9 m/s
Estimated FWHM (@1500 K): 18.8 m/s

- Detected nanoparticles of: ~500.000 AMU
- C_{60} : 720 AMU
- Assuming the same density, the detected nanoparticles' dimensions are:
  ~6 nm!!!
- Assuming polarizability the same as C_{60} then measured density is: ~10^{12} cm^{-3}

<u>For the first time</u> detection of nanoparticles of dimensions of <10 nm, *<u>in situ</u>* in plasma.

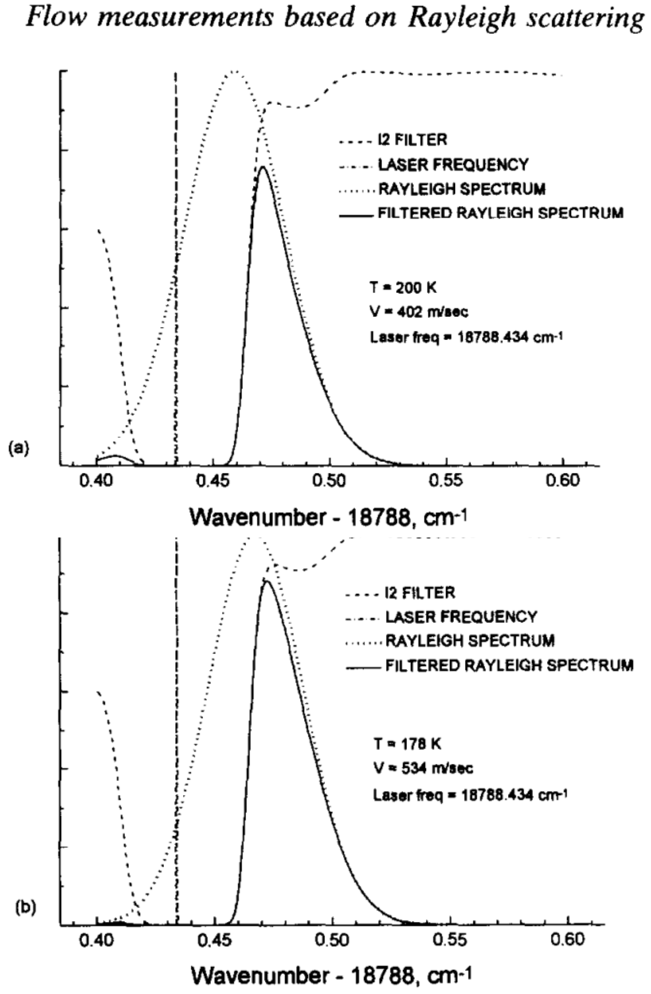
- **Now we are able to map out in situ nanoparticle nucleation and growth in the arc discharge, with high spatial resolution (~100 μm).**



A. Gerakis, Y-W. Yeh, M. N. Shneider, J. M. Mitrani, B. C. Stratton, and Y. Raitses, "Four-Wave-Mixing Approach to *In Situ* Detection of Nanoparticles", Phys. Rev. Applied 9, 014031 (2018)

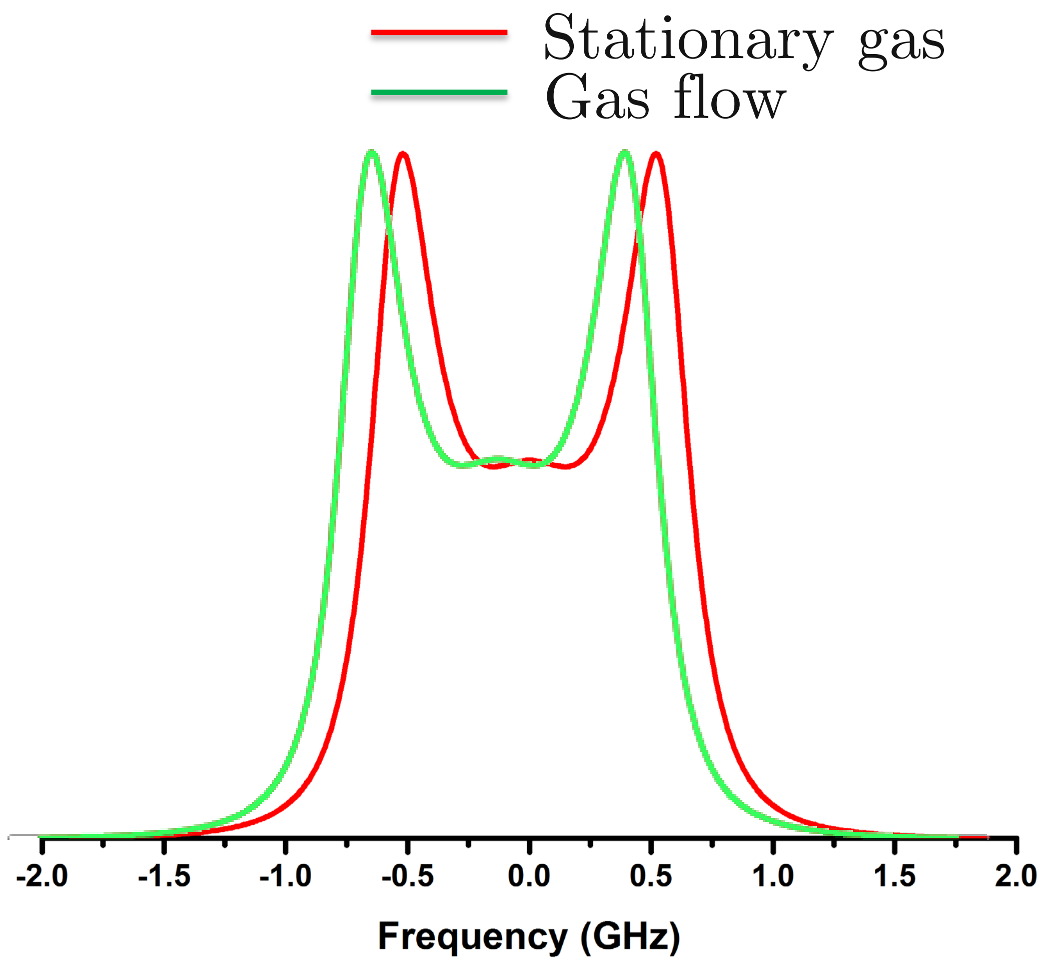
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Neutral gas flow measurement



Filtered Rayleigh scattering

From R. G. Seasholtz, A. E. Buggele, M. F. Reeder, Flow measurements based on Rayleigh scattering and Fabry-Perot interferometer, Optics and Lasers in Engineering, Vol. 27, **6**, 543-570, (1997)



Estimated velocity resolution of 5-10 ms^{-1} in atmospheric or lower pressure (gas pressures down to ~0.1 Torr)

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COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Neutral gas flow measurement

$$\Delta f_{Doppler} = {}^{2 \cdot v_{flow}}/_{\lambda_{probe}}$$

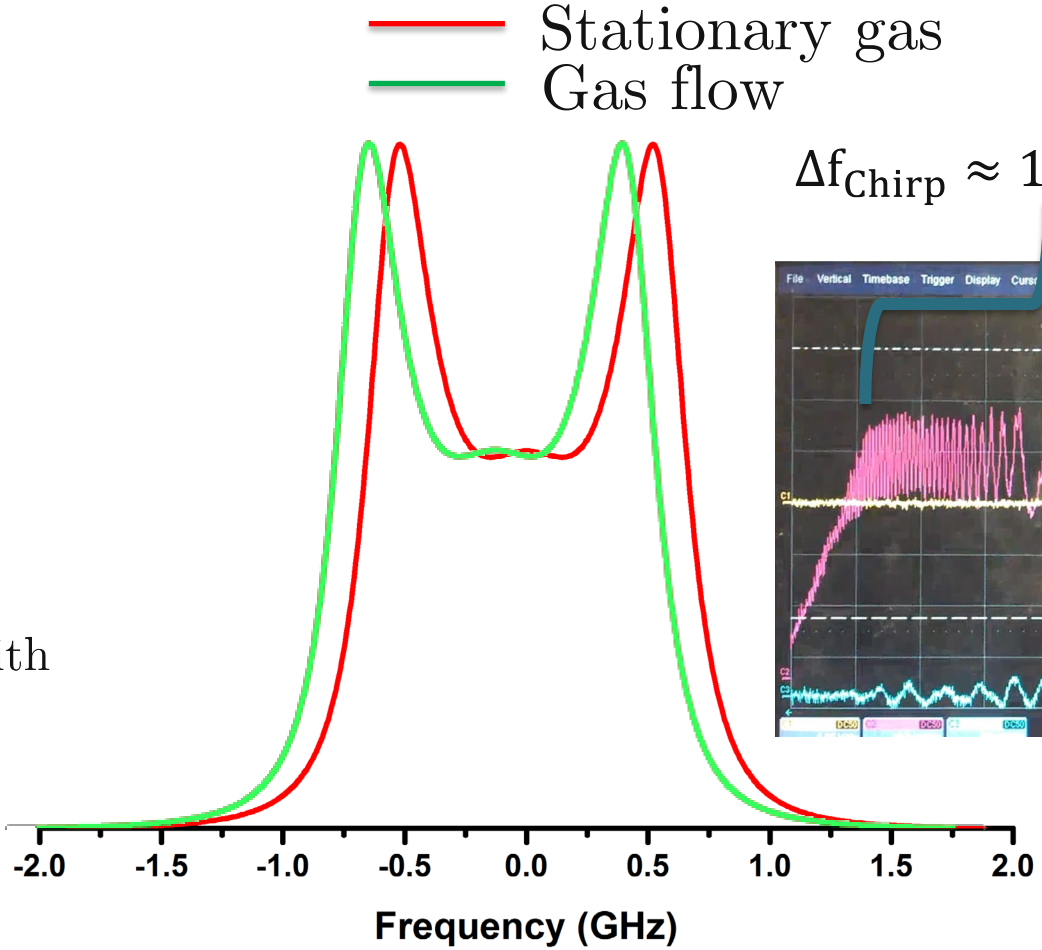
$$v_{flow} = 10\ \mathrm{m} \cdot s^{-1} \quad \lambda_{probe} = 1064\ \mathrm{nm}$$

$$\Delta f_{Doppler} \approx 20\ \mathrm{MHz}$$

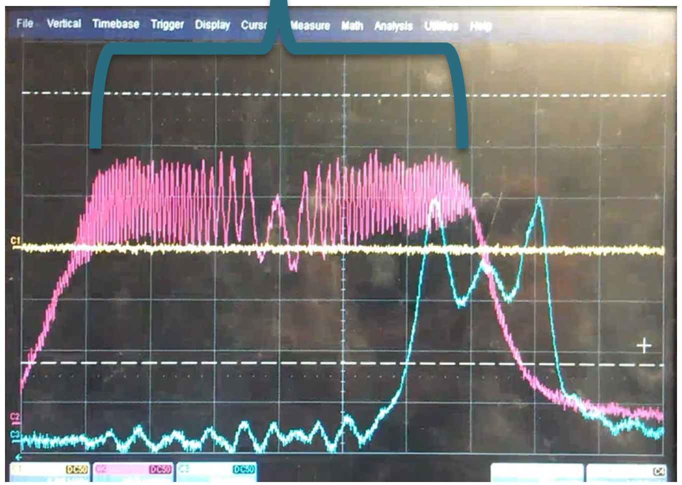
$$\Delta t_{Doppler} \approx 2\ \mathrm{ns}$$

Using a photodiode with 1 GHz bandwidth

Estimated <u>minimum velocity resolution of ~5 ms⁻¹</u> in atmospheric or lower pressure (gas pressures down to ~0.1 Torr @300K)



$$\Delta f_{Chirp} \approx 1.5\ \mathrm{GHz}/150\ \mathrm{ns}$$



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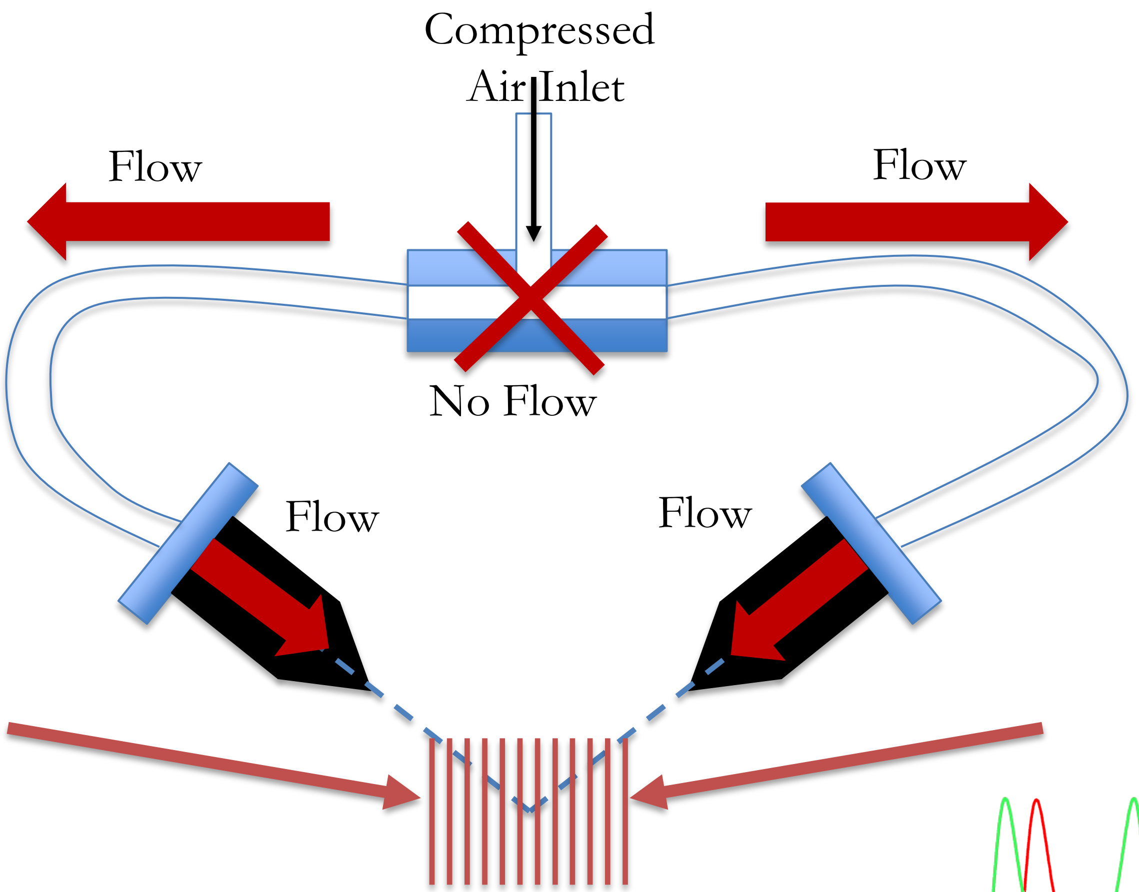
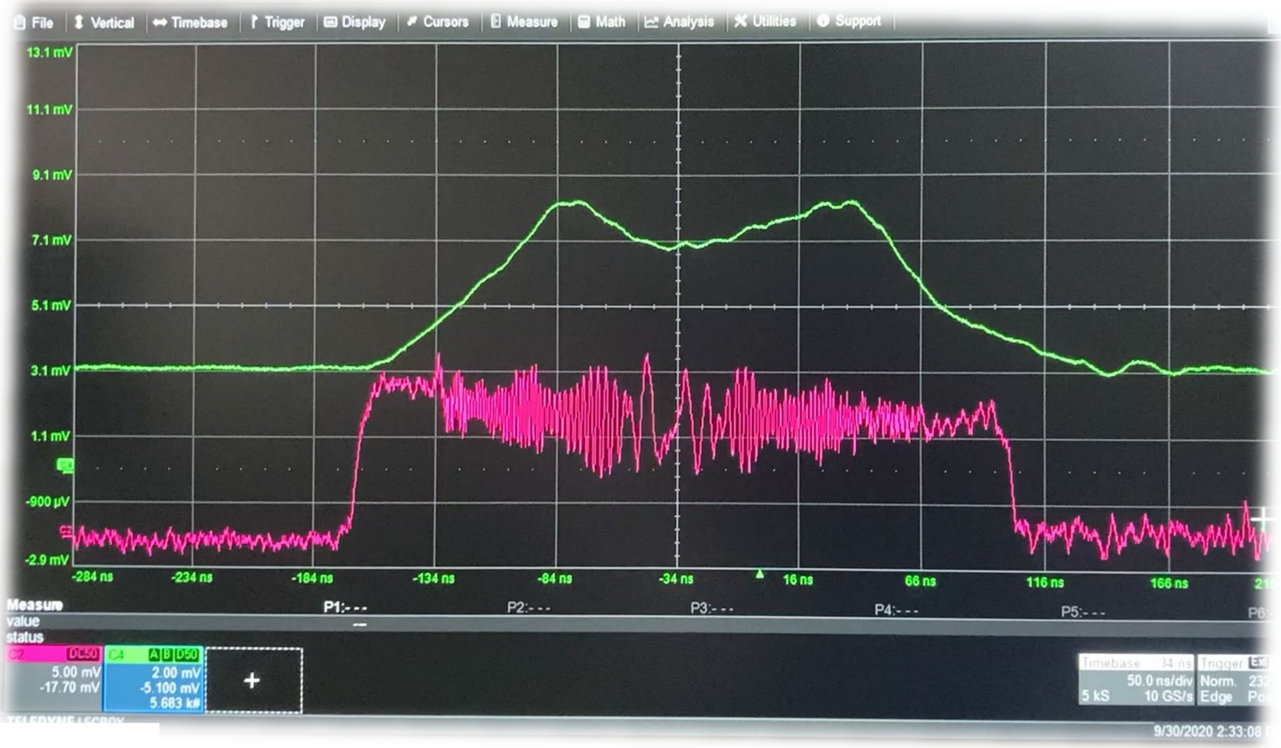
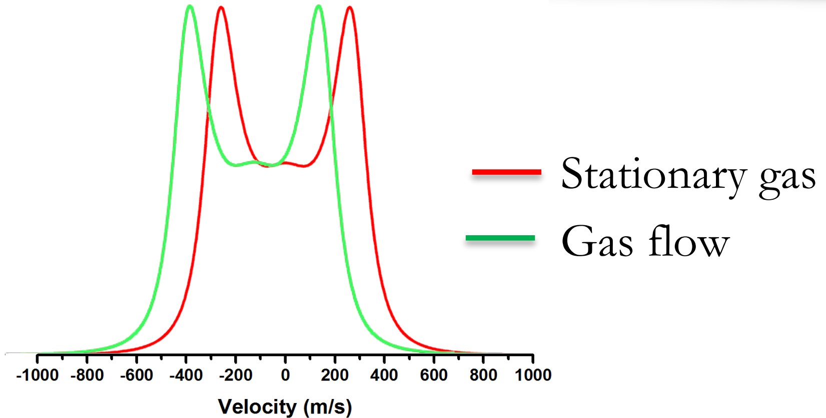
COHERENT RAYLEIGH-BRILLOUIN SCATTERING
Neutral gas flow measurement







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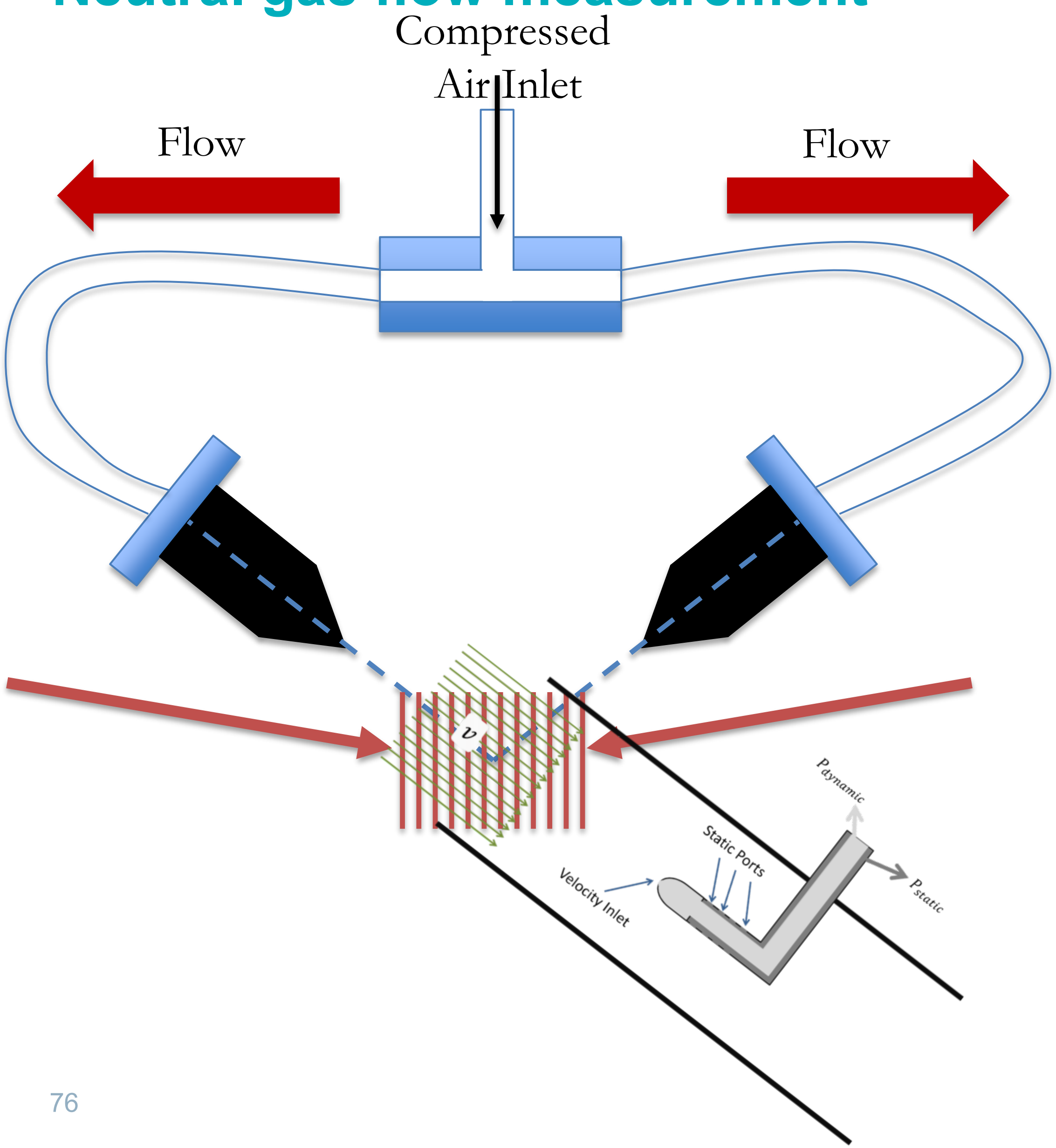
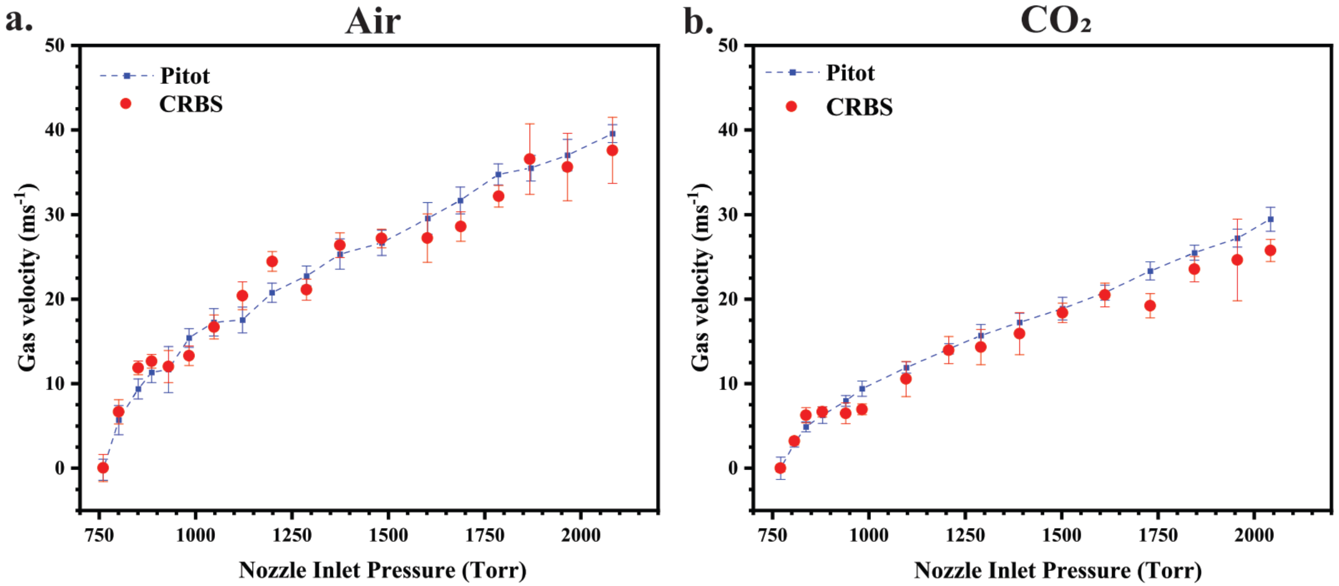
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Neutral gas flow measurement





Alexandros Gerakis, Junhwi Bak, Robert Randolph and Mikhail N. Shneider. "Demonstration of single shot laser velocimetry with coherent Rayleigh-Brillouin scattering," AIAA 2021-0224. *AIAA Scitech 2021 Forum*. January 2021.

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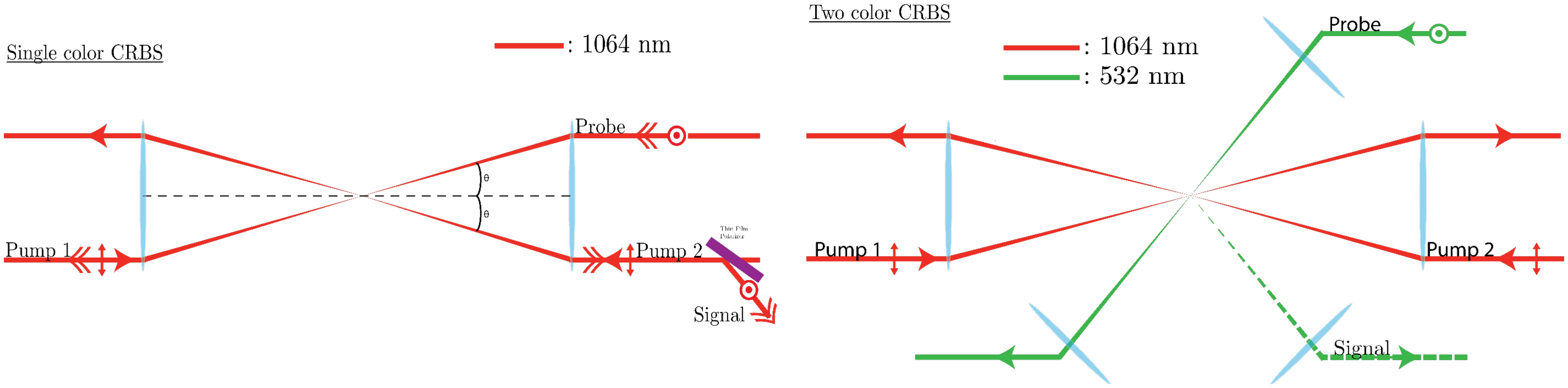
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Two color CRBS scheme



Two-color CRBS scheme

- ➢ Better Signal/Noise Ratio
- ➢ Lower gas densities accessible
  - ❖ Better detector technology @ 532 nm
  - ❖ Spectral filtering of pumps @ 1064 nm
  - ❖ More efficient Rayleigh scattering ($\propto \lambda^{-4}$)

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COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Neutrals in glow discharge measurement

- Measure simultaneously neutral density and translational temperature in glow discharge
  - ➢ Two color, single shot CRBS as means of measurement
  - ➢ Assume ionization ratio small (10^{-6} – 10^{-4}) -> neutrals dominant
  - ➢ Scan radially across the discharge.

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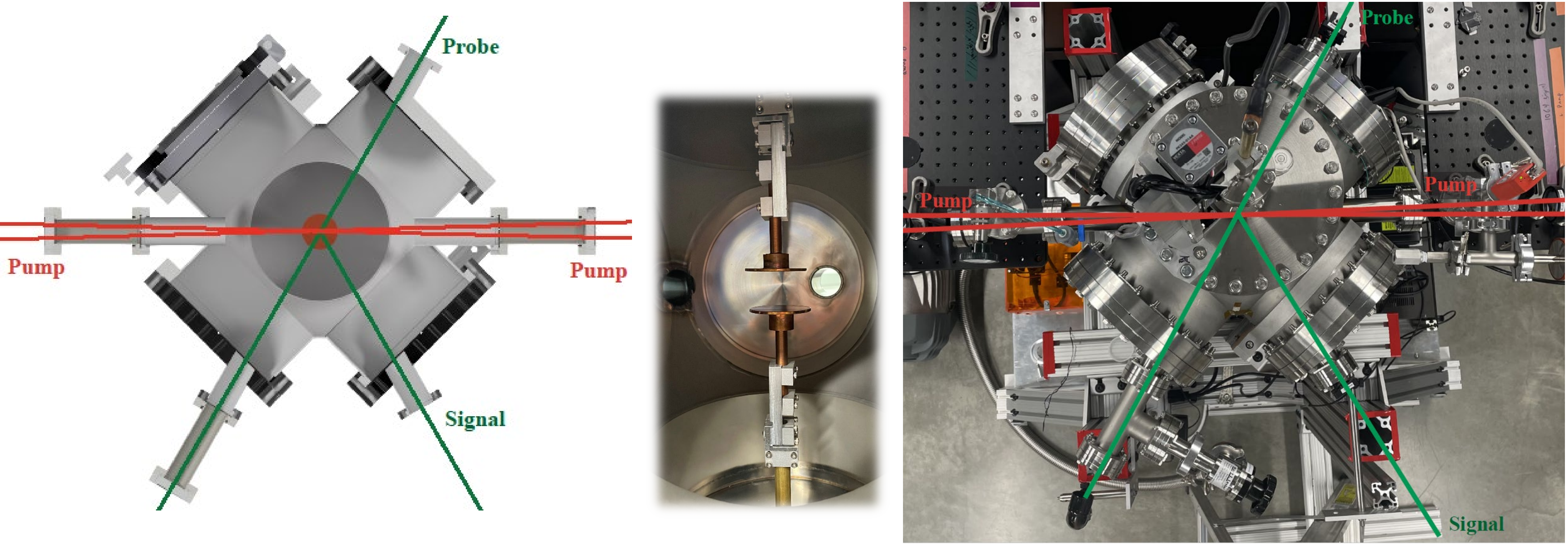
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Neutrals in glow discharge measurement



Dual color CRBS chamber setup

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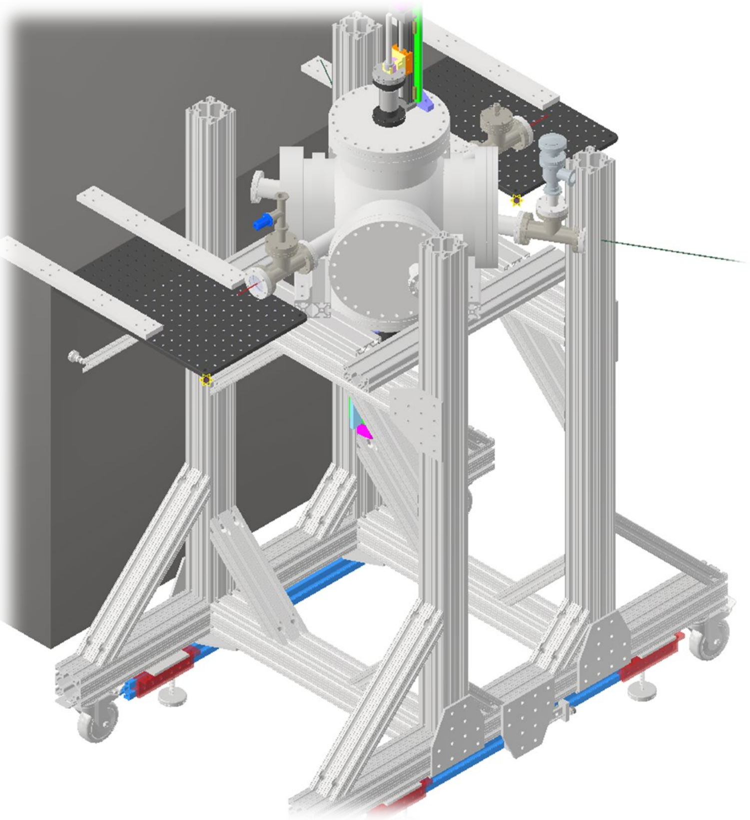
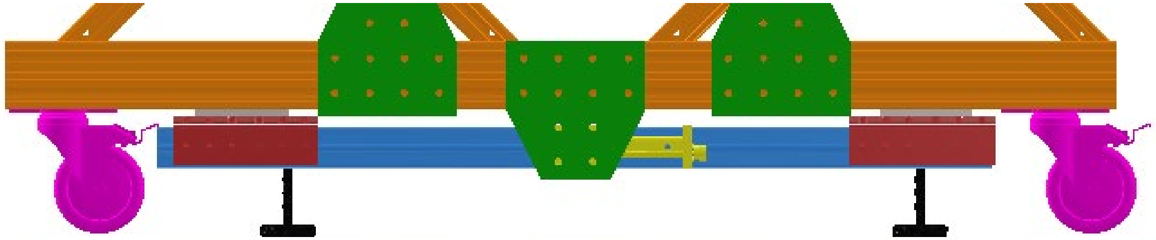
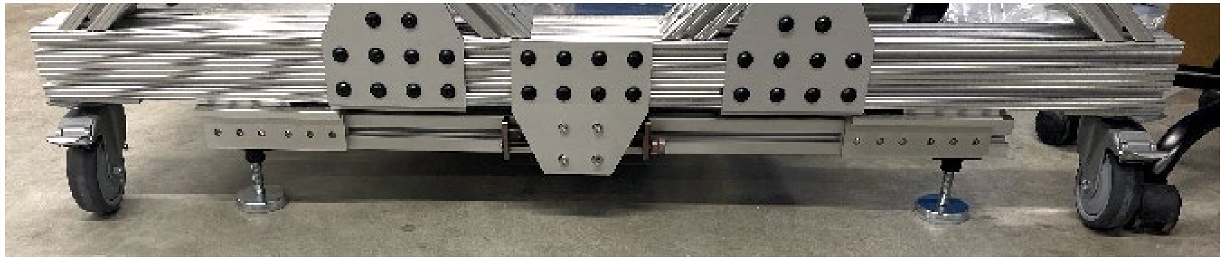
Neutrals in glow discharge measurement







- Measurements radially across the positive column of glow discharge, from center to periphery.
  - ➢ Assumption: Positive column is radially symmetric
  - ➢ Assumption: Operating conditions are the same from point to point and run to run.

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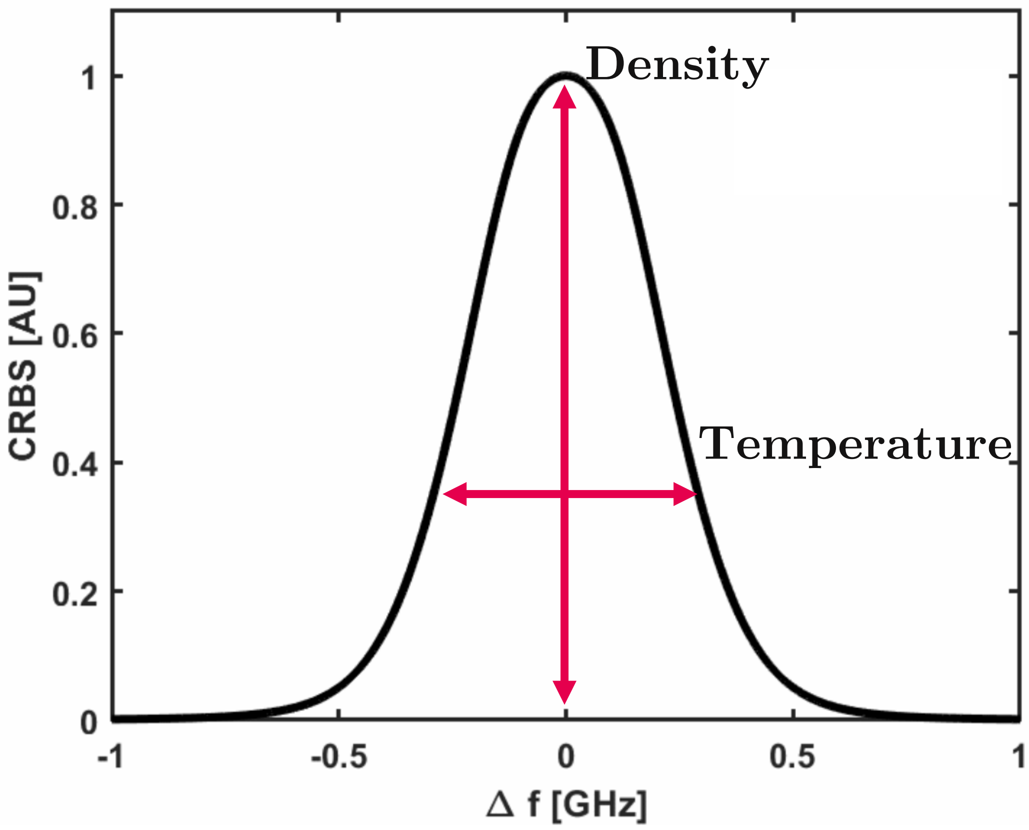
COHERENT RAYLEIGH-BRILLOUIN SCATTERING

Neutrals in glow discharge measurement



Xenon 15 Torr @300K

- **Temperature:**

$$f(v_z) \propto e^{\frac{-mv_z^2}{2kT}}$$

- **Density:** $I_S \propto \Delta n^2$

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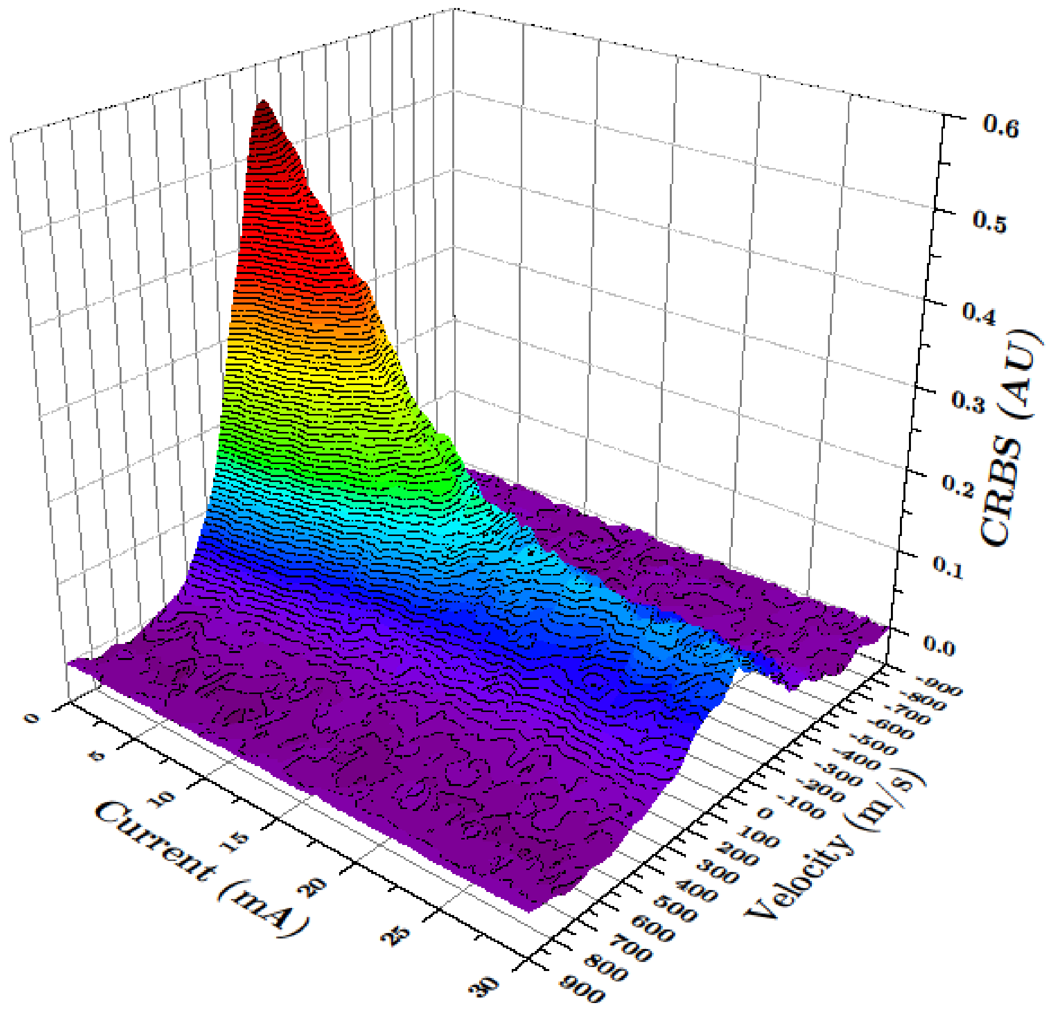
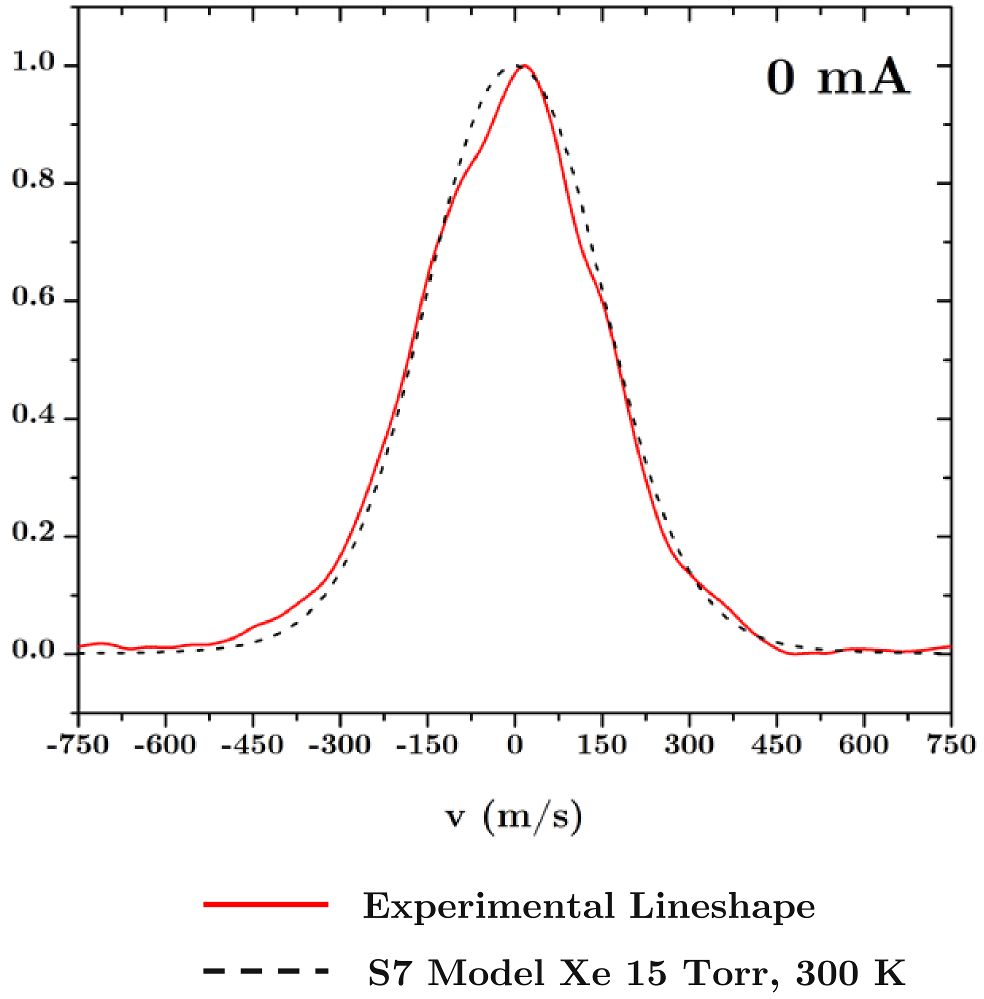
Neutrals in glow discharge measurement





Pan, Xingguo & Shneider, M.N. & Miles, R.B.. (2004). Coherent Rayleigh-Brillouin scattering in molecular gases. Physical Review A. 69. 33814-. 10.1103/PhysRevA.69.033814.

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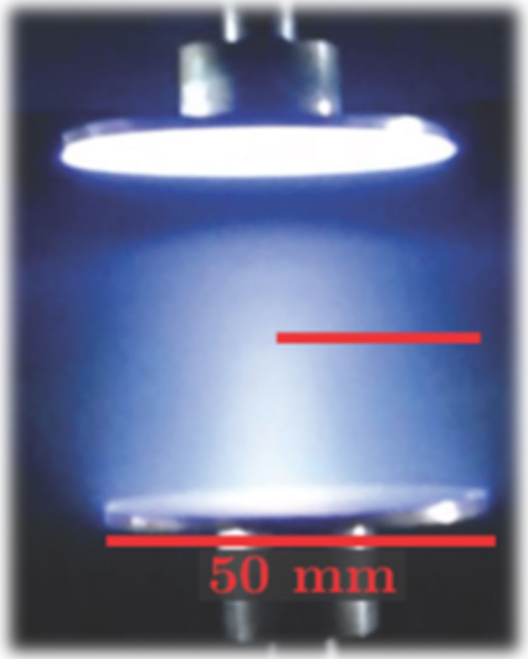
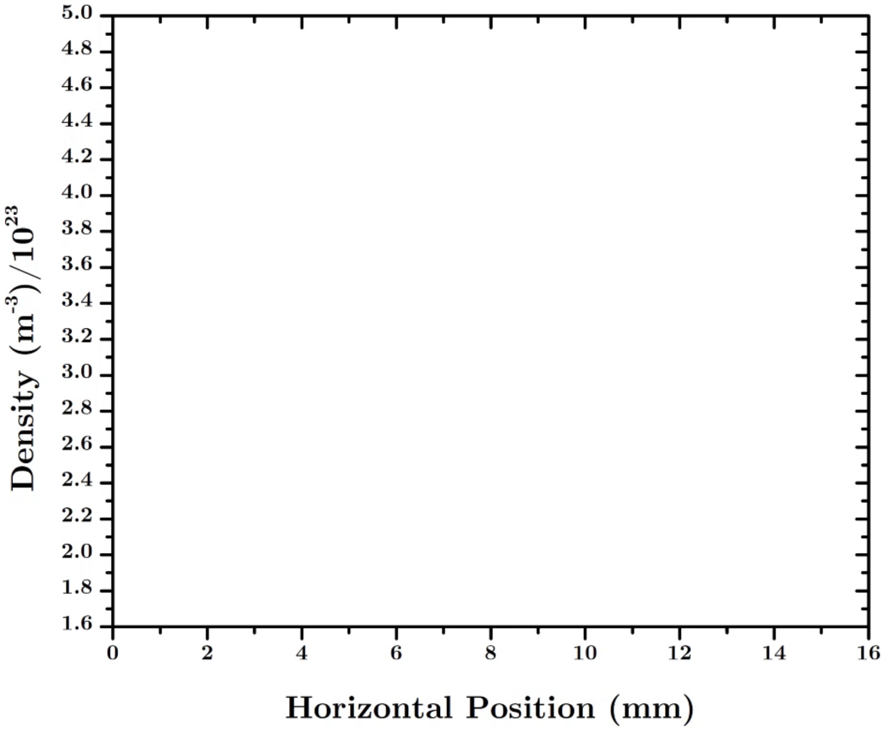
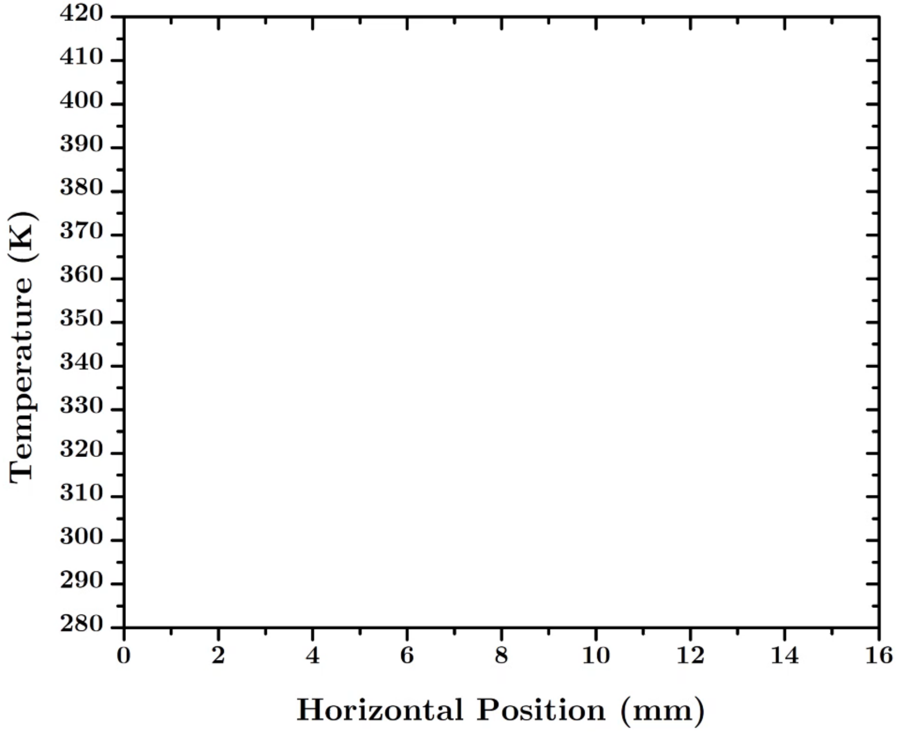
Neutrals in glow discharge measurement







Glow in 15 Torr Xe

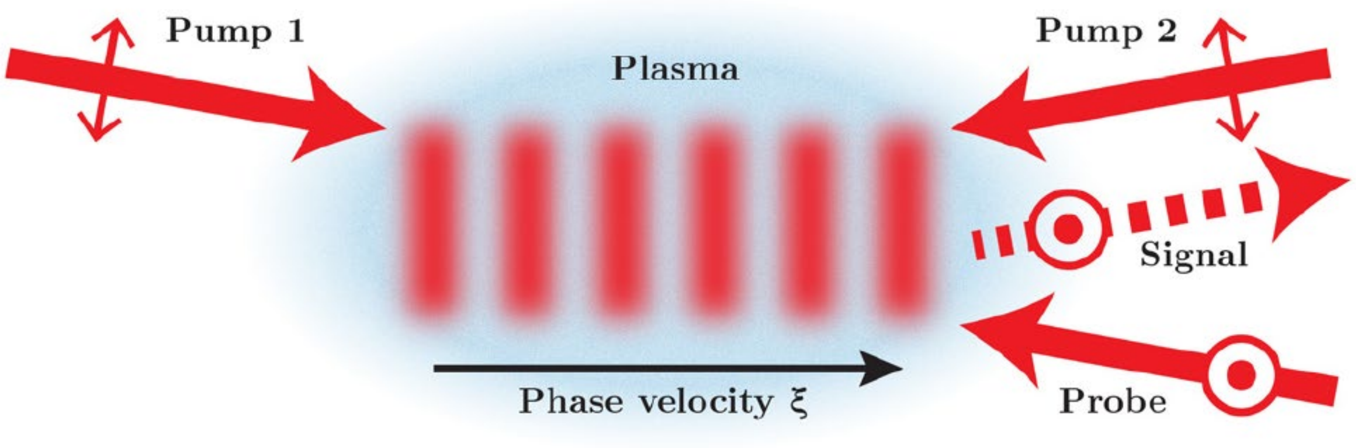
- Average of 100 samples taken at each point, samples collected at 10 Hz rate

(single spectrum acquisition time ~150 ns)

	Density (10^{-23} m^{-3})	Temperature (K)
Edge	4.8$\pm$ 0.4	295$\pm$ 15
Center	1.9$\pm$ 0.4	370$\pm$ 15

COHERENT THOMSON SCATTERING

A novel way to measure electron temperature?



Physics of Plasmas ARTICLE scitation.org/journal/php

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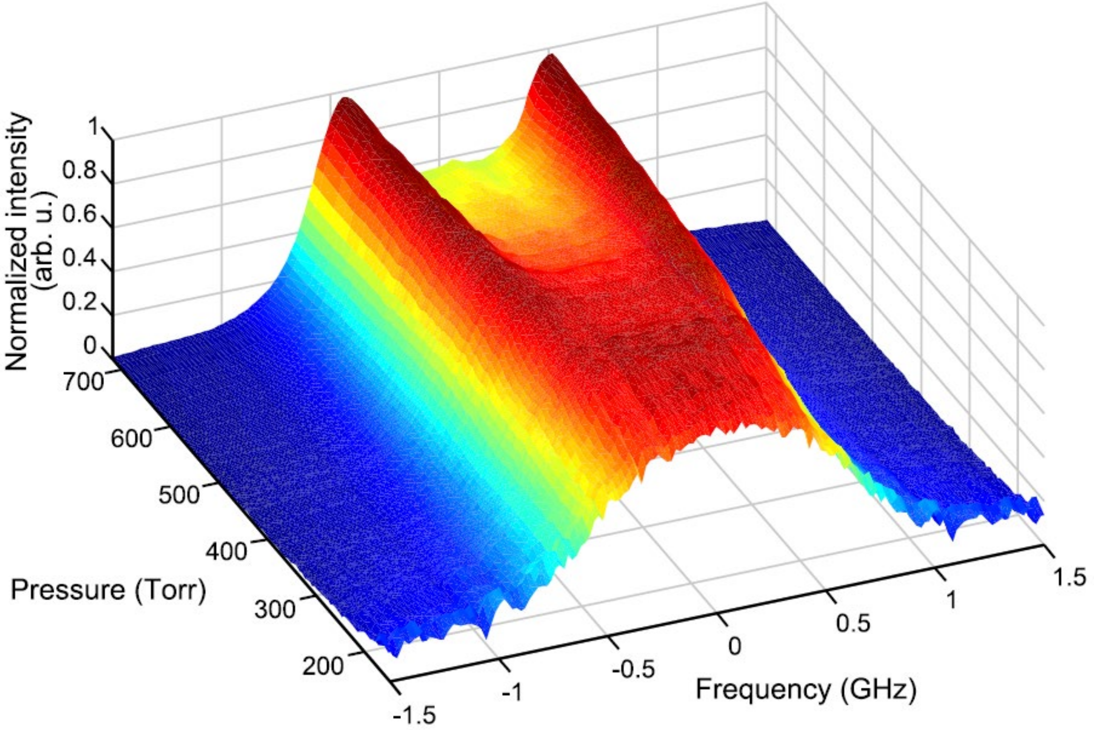
LASER DIAGNOSTICS
Summary

- There's no "one-size fits all": different techniques are complimentary towards the solution of the same problem.
- Ideal way to characterize plasma & plasma flow measurements.
- Work with (almost) every gas.
  - Advantage <u>and</u> disadvantage
- Time to replace all mechanical probes ☺



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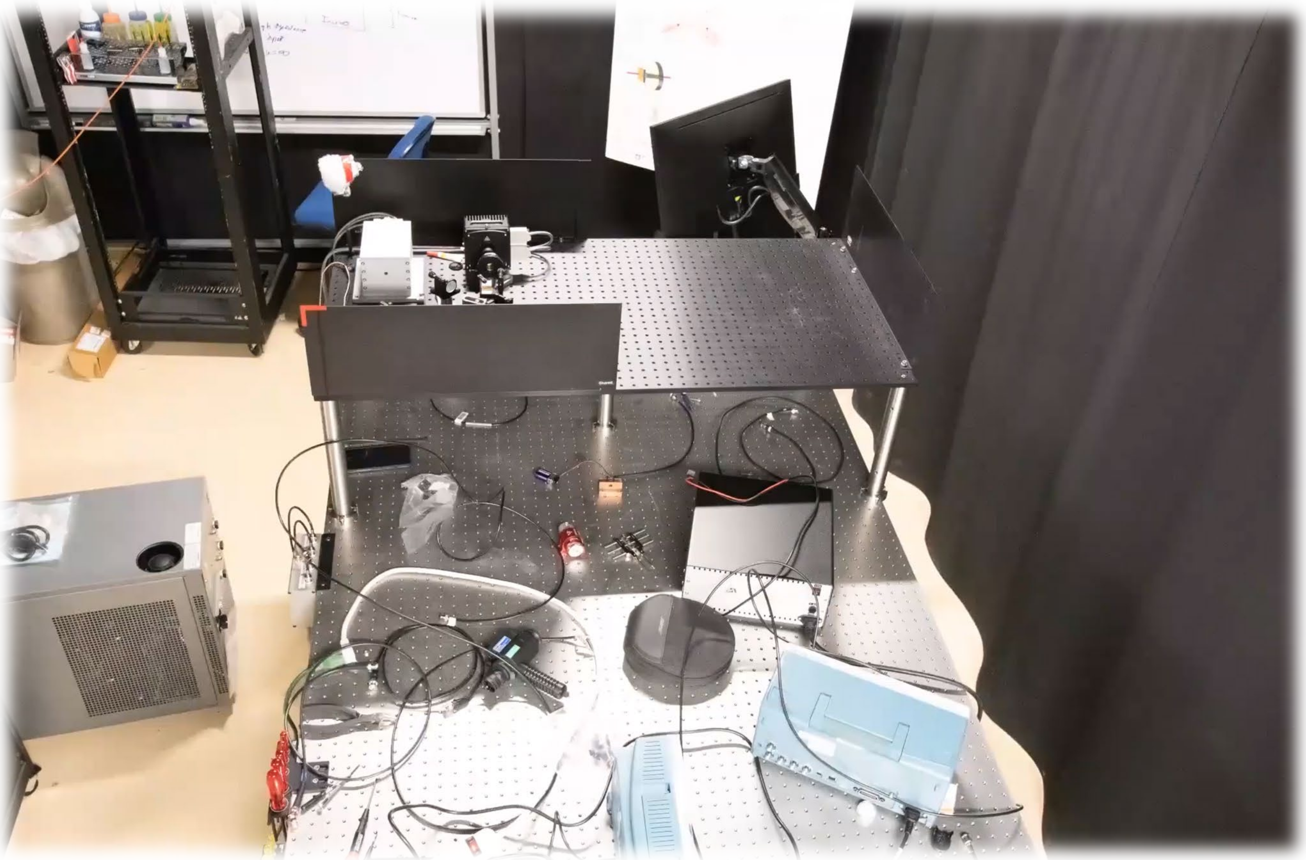

Thank you!
Questions?

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