

# Single-bubble Sonoluminescence



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# Outline

- Background
  - What is sonoluminescence?
  - History
- Bubble Motion
  - Radial and translational
- Parameters
  - Fluid temperature, bubble composition, etc.
- Spectra
  - Surrounding fluid and bubble
- Theories
- Conclusions

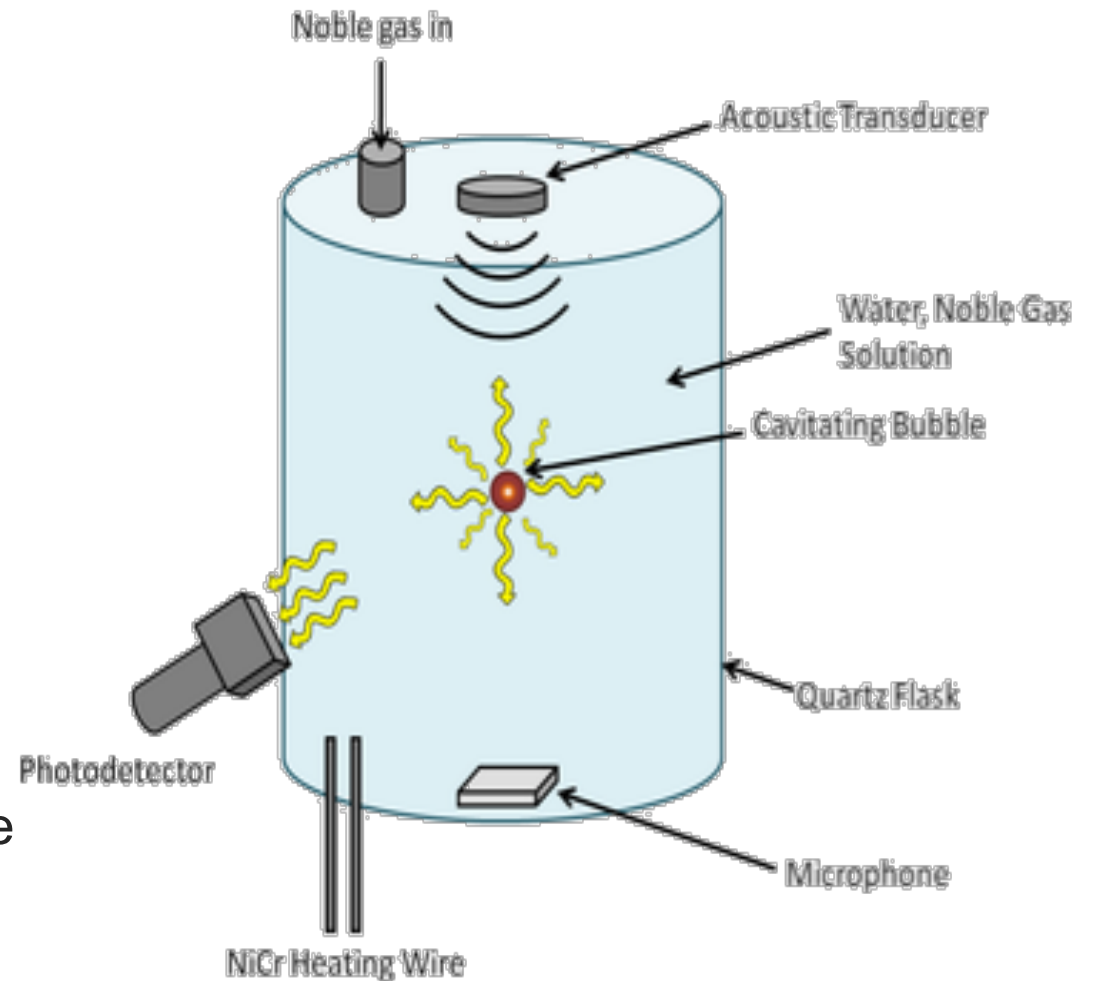


Diagram of sonoluminescence apparatus

# Background-What is Sonoluminescence?

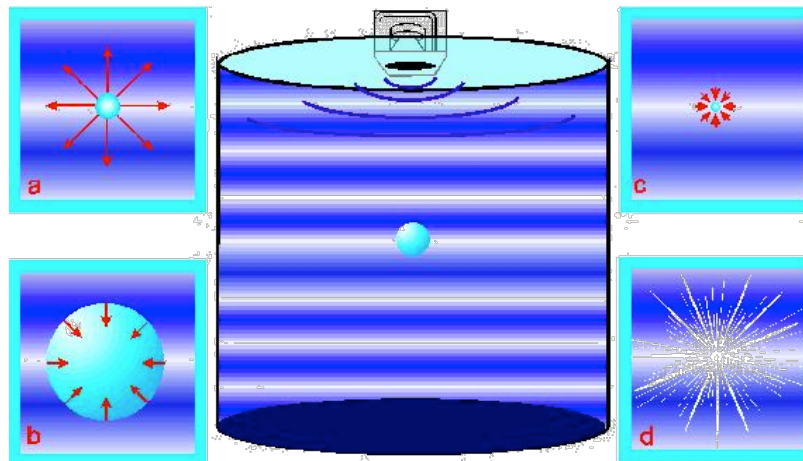
- Sonoluminescence is visible light emitted from an acoustically driven bubble
- Bubble oscillates non-linearly in micrometer range
- The average temperature of the bubble at collapse is  $\sim 10^3$  K
- Light is emitted in picosecond pulse widths depending on bubble composition
- Sonoluminescence has been maintained continuously for billions of cycles
- Sonoluminescence is producible in many fluids



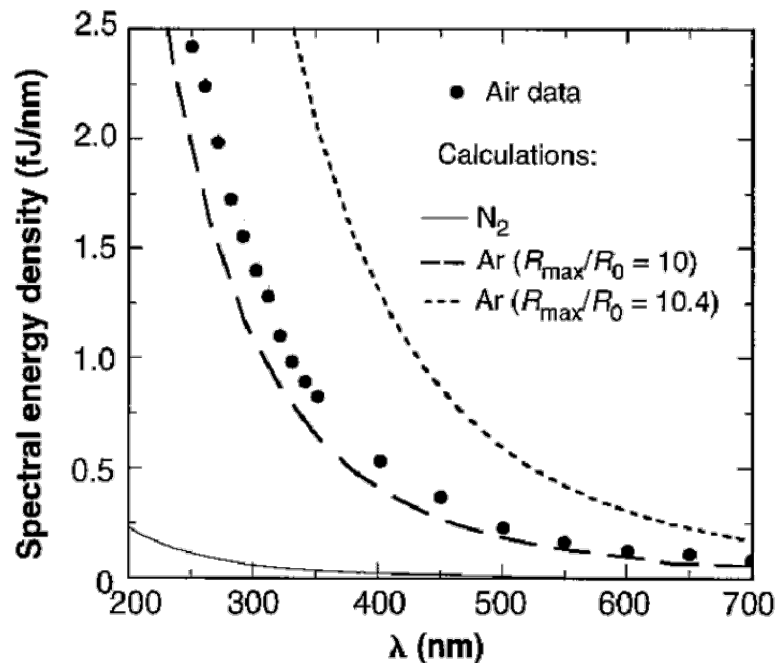
Sonoluminescent bubble

# Background-What is Sonoluminescence?

- Acoustically driven  
0.5-50  $\mu\text{m}$  radius  
bubble oscillates  $\sim 20$  kHz
- Light is usually emitted in 100-200 picosecond window around minimum radius
  - Composition of bubble effects pulse width
- Center of bubble believed to be hottest
- Requires non-reactive molecules
  - Typically noble gases
- Spectral lines visible for low enough driving frequency and depending on fluid used



- Bubble expands from equilibrium radius  $\sim 5$  micrometers
- Bubble expands to maximum radius of  $\sim 50$  micrometers
- Bubble collapses to minimum radius  $\sim 0.5$  micrometers
- Bubble emits 100-200 picosecond pulse of light



- Nitrogen is relatively non-reactive
- Air contains argon and trace amounts of other noble gases

# Background-History

- Late 19<sup>th</sup> century – Lord Rayleigh first studied cavitation
  - Work led to Rayleigh-Plesset Equation
- 1934 - multi-bubble sonoluminescence discovered by Frenzel and Schultes
- 1989 - single-bubble sonoluminescence discovered by Gaitan
- 1994 - Hiller *et al.* discovered dependence of light emission on type of gas in bubble



Image of a collapsing bubble

# Navier-Stokes Equation

- Models fluid motion
- Non-linear partial differential equation
  - From Newton's second law and (classical) conservation laws
- Comes from conservation of momentum, energy and mass
- Rayleigh-Plesset equation is derived from this
  - Used to solve for radial motion of bubble

$\rho$  - fluid density

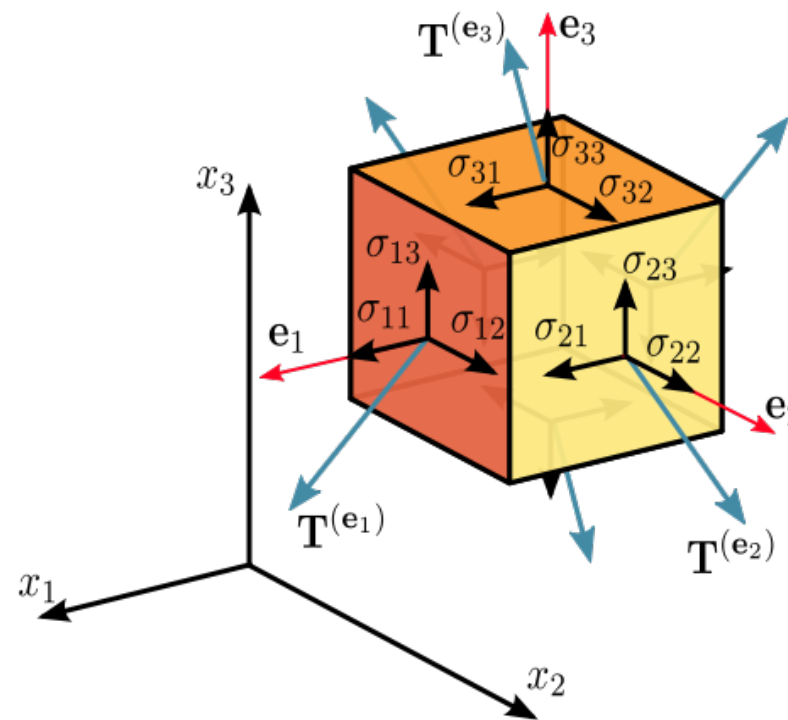
$\mathbf{v}$  - fluid velocity

$p$  - pressure

$\mathbf{T}$  - stress deviator tensor

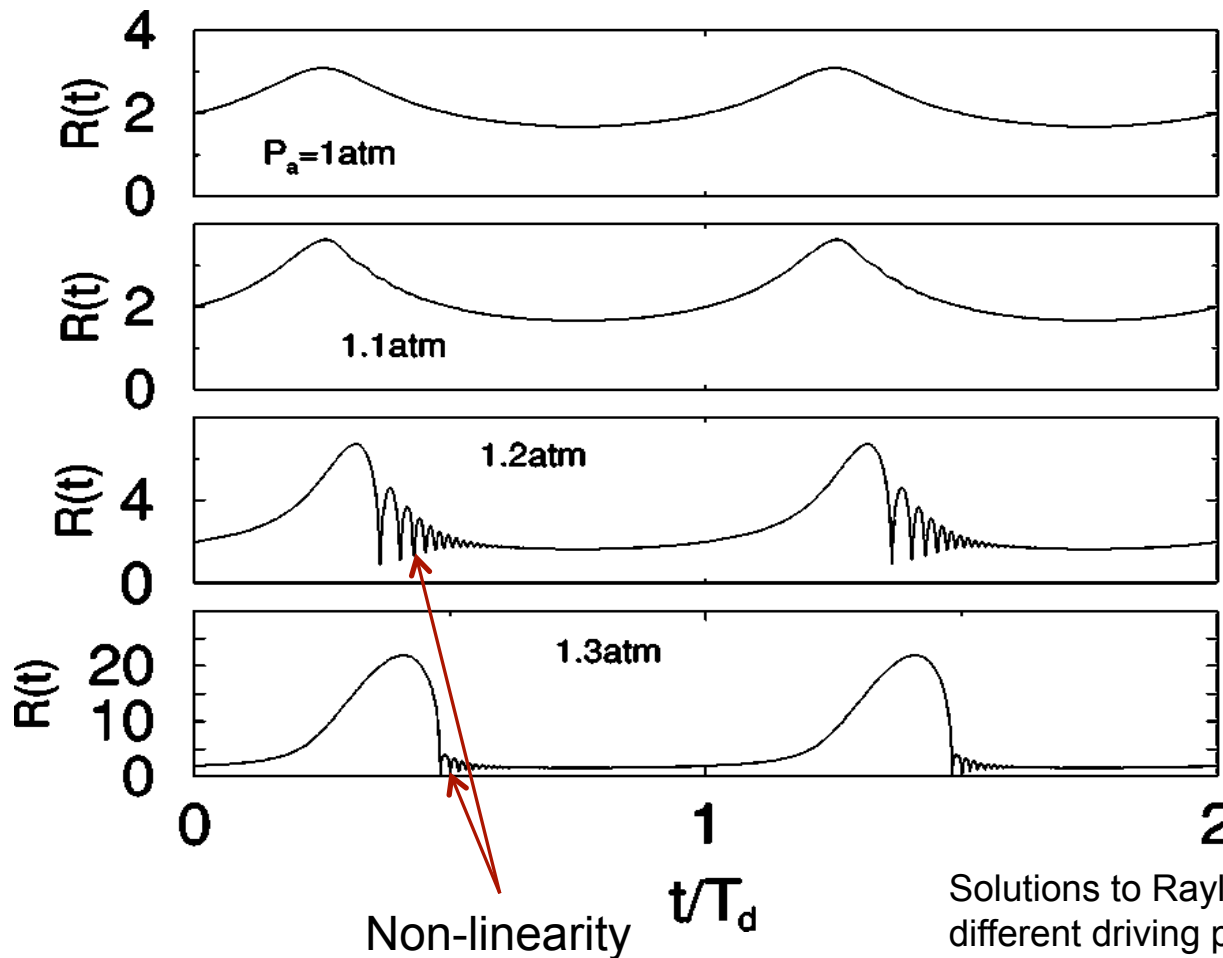
$\mathbf{f}$  - force density

$$\rho \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + \nabla \cdot \mathbf{T} + \mathbf{f}$$



# Rayleigh Plesset Equation

$$\rho \left( R\ddot{R} + \frac{3}{2}\dot{R}^2 \right) = \left[ p_g(R,t) - P_0 - P(t) \right] - 4\mu \frac{\dot{R}}{R} - 2\frac{\sigma}{R} + \frac{R}{c} \frac{d}{dt} p_g$$

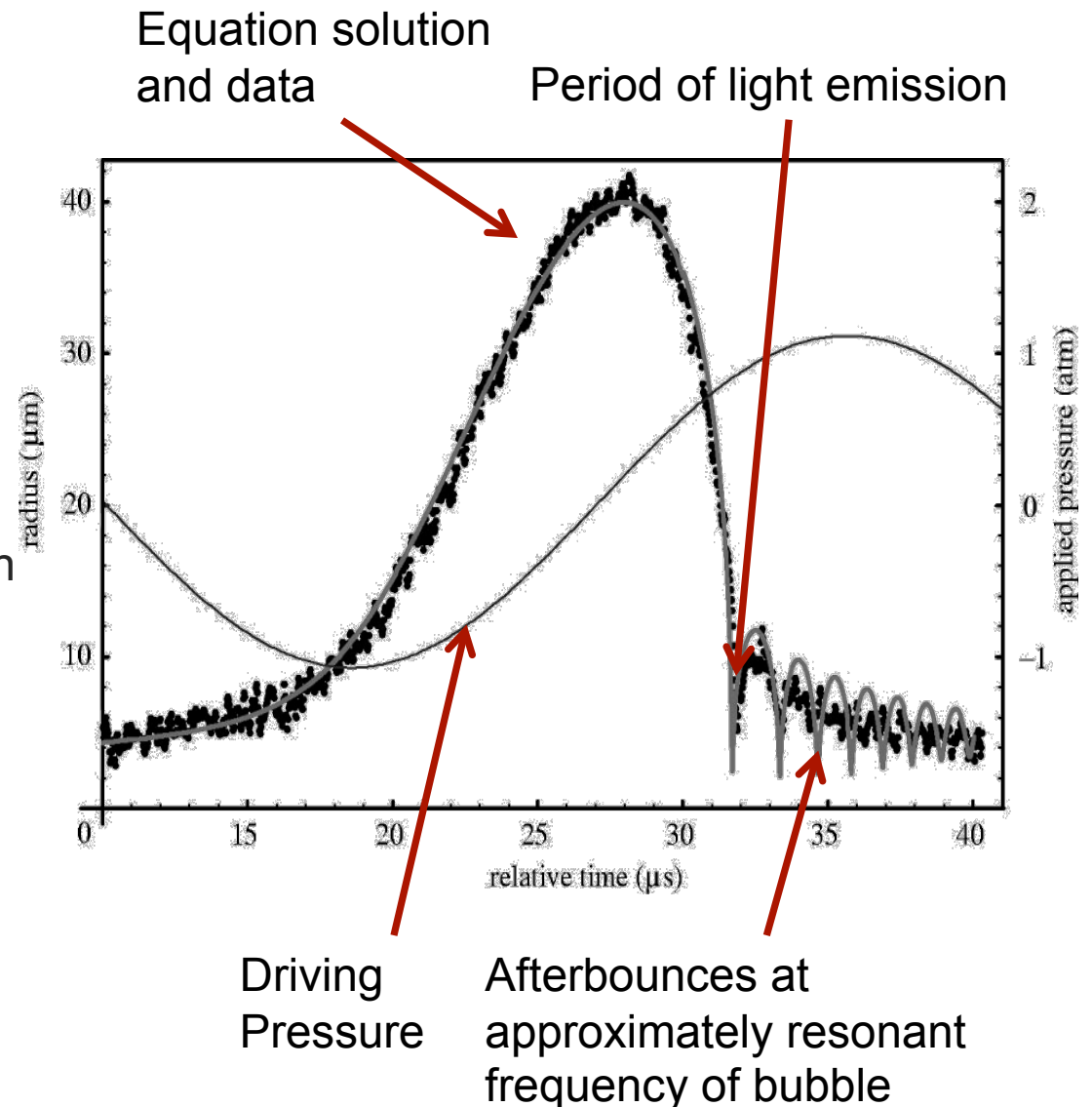


$\rho$  – density of liquid  
 $R$  – bubble radius  
 $p_g$  – pressure in bubble  
 $P_0$  – pressure far from bubble  
 $P(t)$  – driving pressure  
 $\mu$  – shear viscosity of liquid  
 $\sigma$  – surface tension at interface  
 $c$  – speed of sound in liquid  
 $h$  – van der Waal's hard core radius  
 $\kappa \approx 1$ , effective polytropic exponent (isothermal)

Solutions to Rayleigh-Plesset Equation for different driving pressures (equilibrium radius, 2  $\mu\text{m}$ ; frequency, 26.5k Hz)

# Rayleigh-Plesset Equation

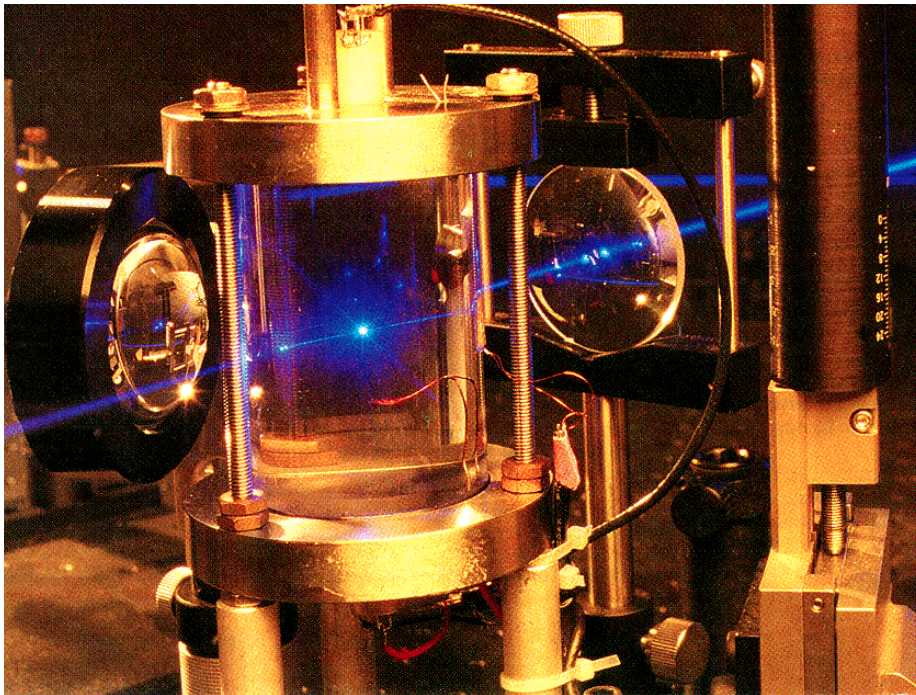
- Rayleigh-Plesset Equation describes motion of spherical bubble
- Small perturbations are allowed but can cause fragmentation if too large
- Approximate surrounding liquid (typically water) as incompressible
- Assume there to be no thin-film
- Extremely sensitive to changes in conditions
- Approximate isothermal expansion and adiabatic compression
- Breakdown when  $\frac{\dot{R}}{c} \sim 1$  and during collapse
- Numerically solvable, large amount of approximation required for analytic solution



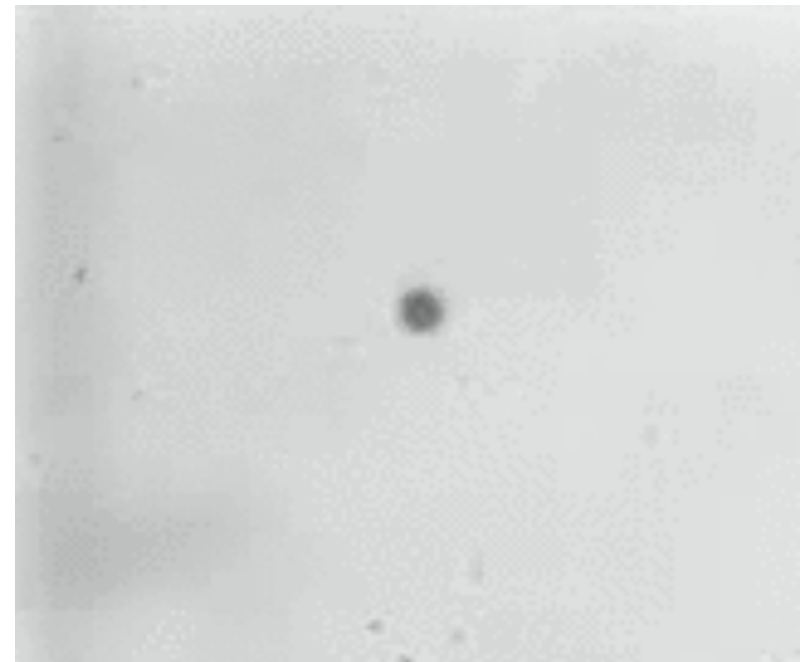


# Measuring Bubble Radius

- Light from laser is incident on the bubble
- Light scattered from bubble is used to determine bubble radius because number of photons that reach photomultiplier is reduced



Laser used to measure radius of bubble



Oscillating bubble

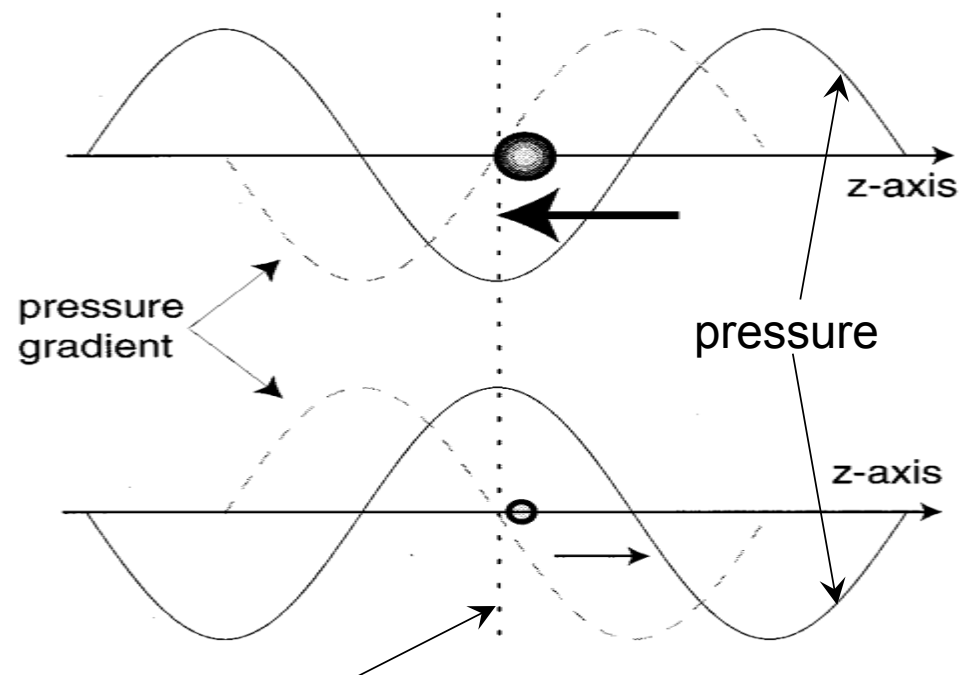
# Translational Motion-Bjerknes Force

- Bubble forced toward pressure antinode (maxima) because resonant frequency of bubble is greater than driving frequency,  $\omega_0 > \omega$
- Bjerknes force due to pressure gradient across bubble
- Bubble position comes from buoyancy and Bjerknes forces

$$F_{buoyancy} = \frac{\rho g}{T} \int_0^{T_d} V(t) dt$$

$$\vec{F}_{Bjerk} = \left\langle \hat{b} \cdot \vec{F}_{bubble} \right\rangle = \left\langle -\frac{4}{3} \pi R(t)^3 \nabla p \right\rangle$$

Pressure gradient is zero in pressure minima or maxima

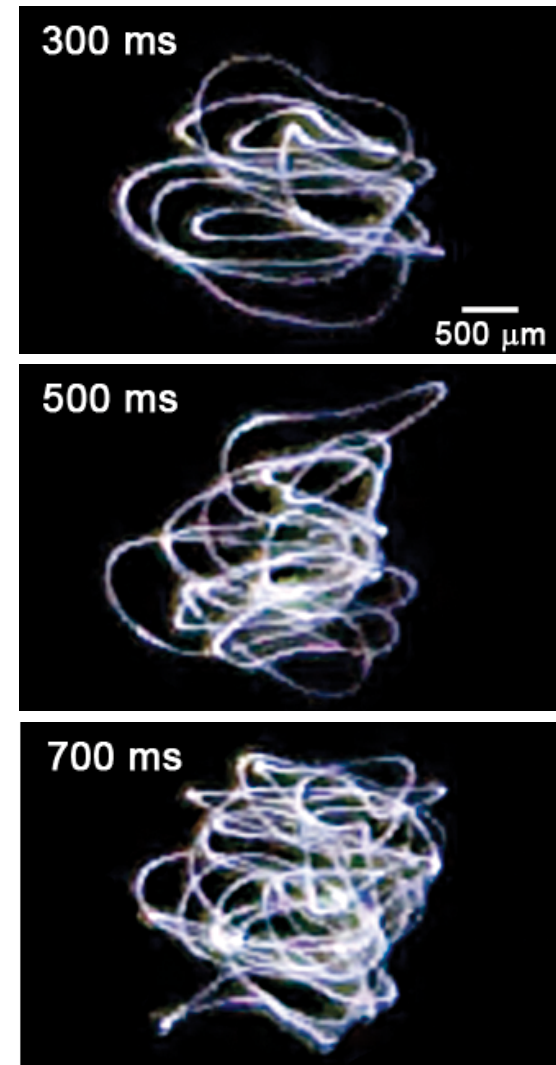


location of pressure antinode

# Bubble Motion

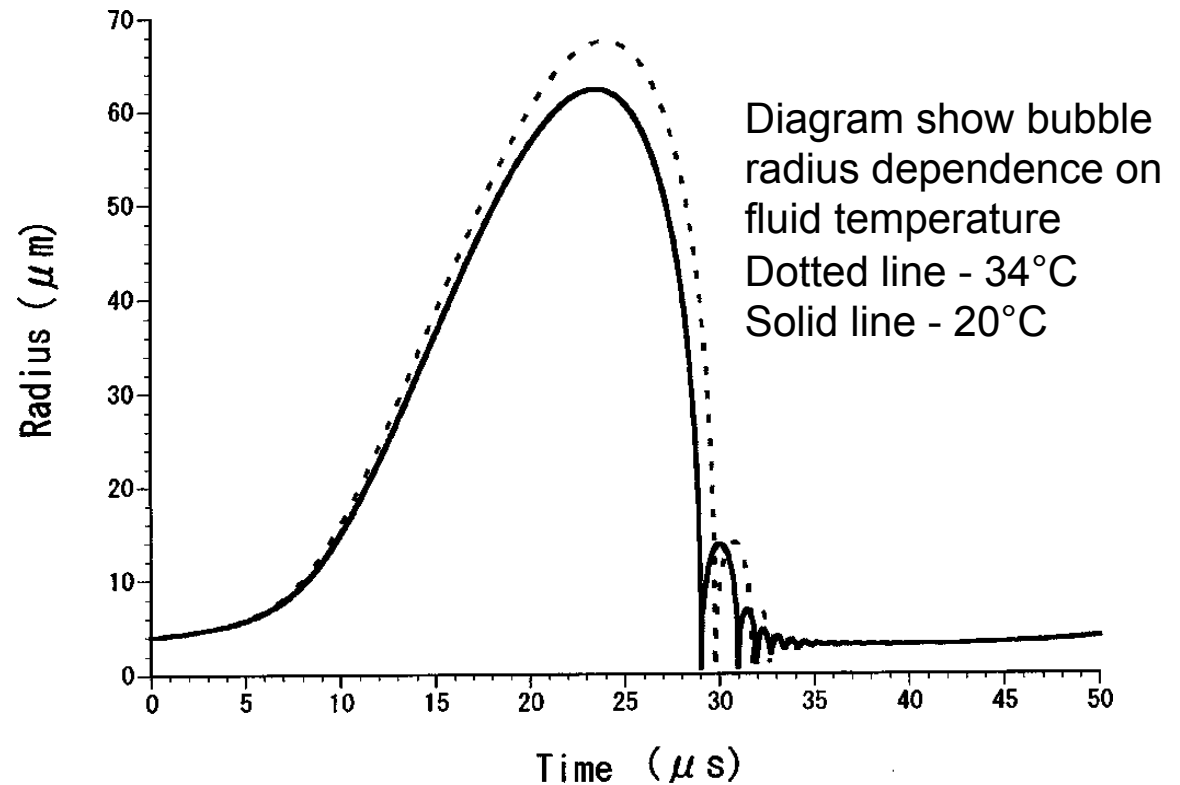
- In simple model, Bjerknes and buoyancy forces are equated, but they are not actually equal
- Bubble has translational motion in addition to radial motion
- Viscosity of fluid influences form of translational motion
  - Lower viscosity fluids (water) have circular paths
  - Higher viscosity fluids ( $\text{H}_3\text{PO}_4$ ) have elliptical paths

Argon bubble in  $\text{H}_3\text{PO}_4$  seen with translational motion



# Surrounding Temperature and Bubble Radius

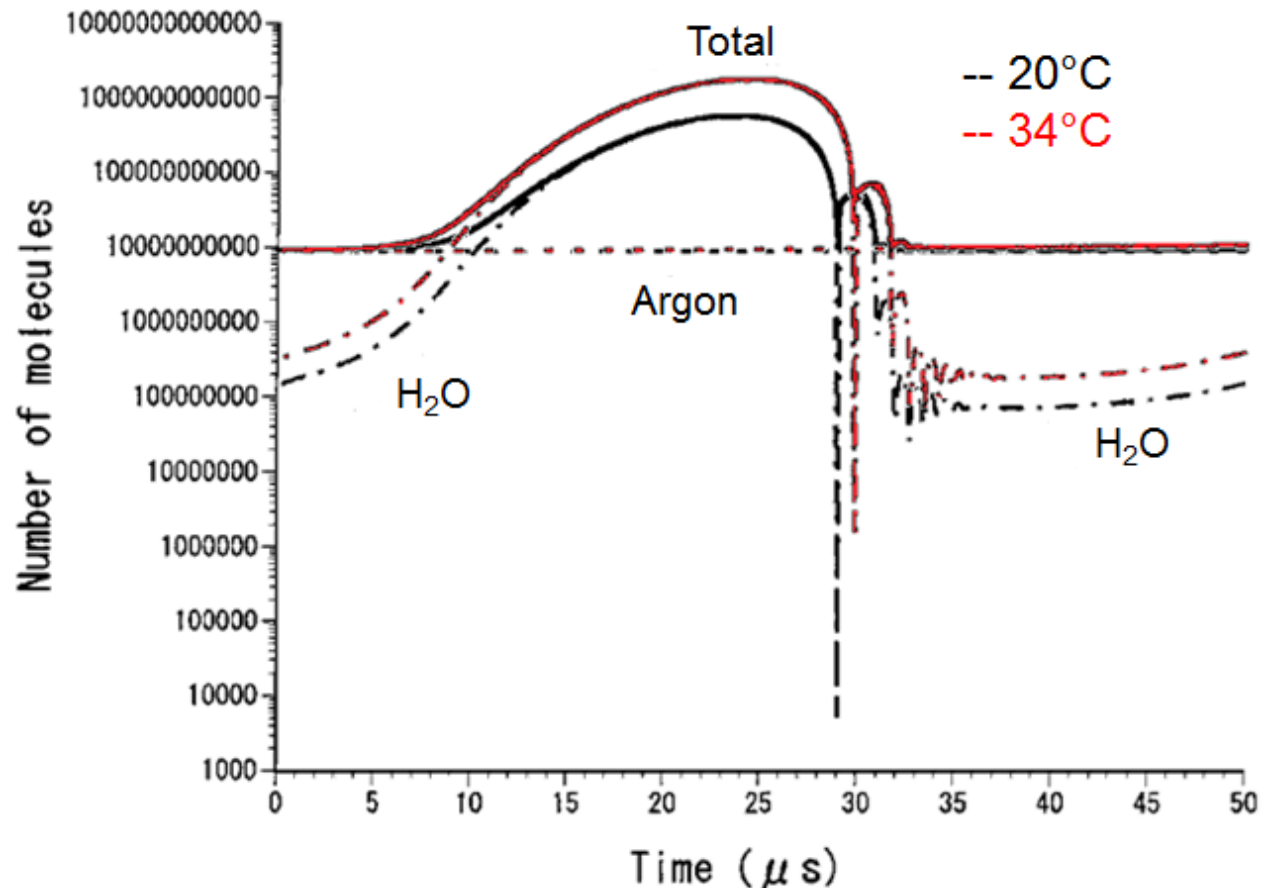
- Surface tension and viscosity constrict bubble expansion
  - Lower surrounding temperature constricts bubble expansion
- Higher surrounding temperature causes more water vapor to enter bubble, causing less light emission



Water Temperature °C	Surface Tension $\sigma \cdot 10^{-3}$ N/m	Viscosity $\mu \cdot 10^{-3}$ Pa·s	Vapor Pressure $p_v \cdot 10^3$ Pa
20	72.8	1.0	2.3
34	70.5	0.74	5.3

# Fluid Temperature and Bubble Composition

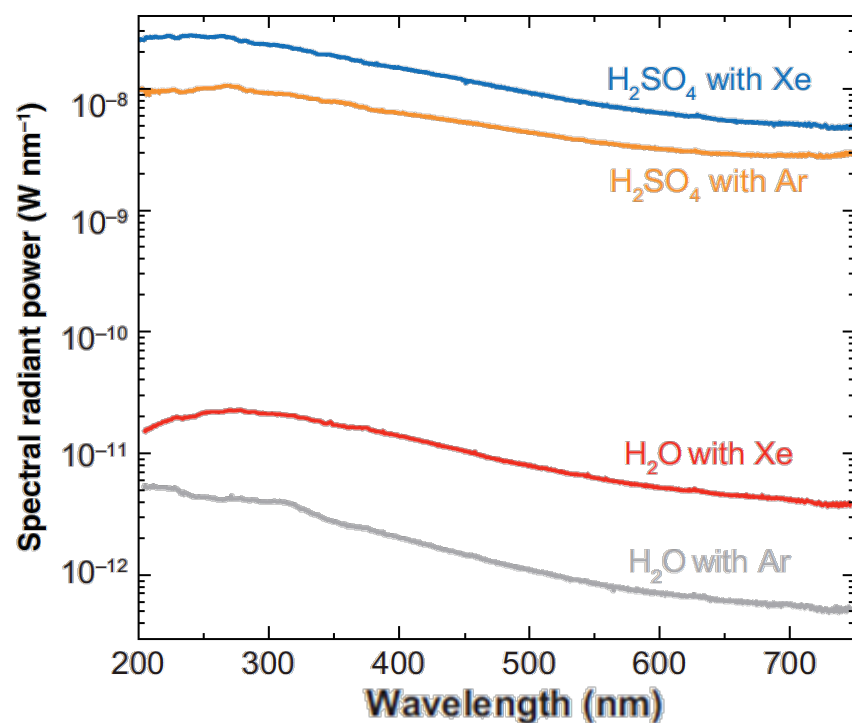
- Pressure from fluid causes influx of molecules when radius is large and outflow when radius is small
- Bubble temperature at collapse is higher in colder fluid because less water vapor is trapped inside bubble at the collapse due to the lower-saturated vapor pressure



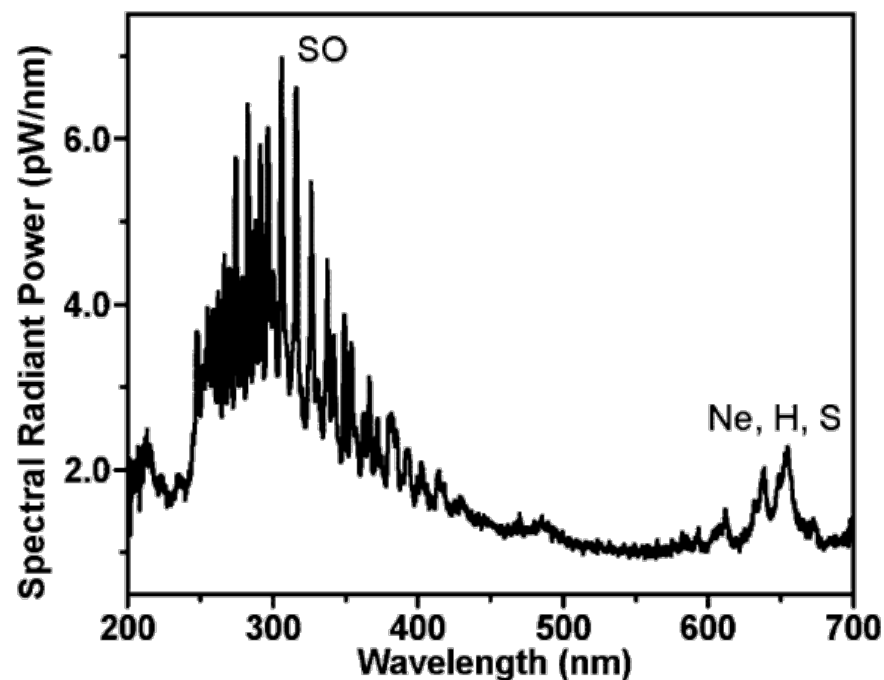
Number of molecules in bubble for one acoustic cycle for two temperatures

# Sonoluminescence Spectrum

- Fluid and noble gas both affect sonoluminescence spectrum
- Vibrational frequencies are visible
- Water has higher specific heat than sulfuric acid



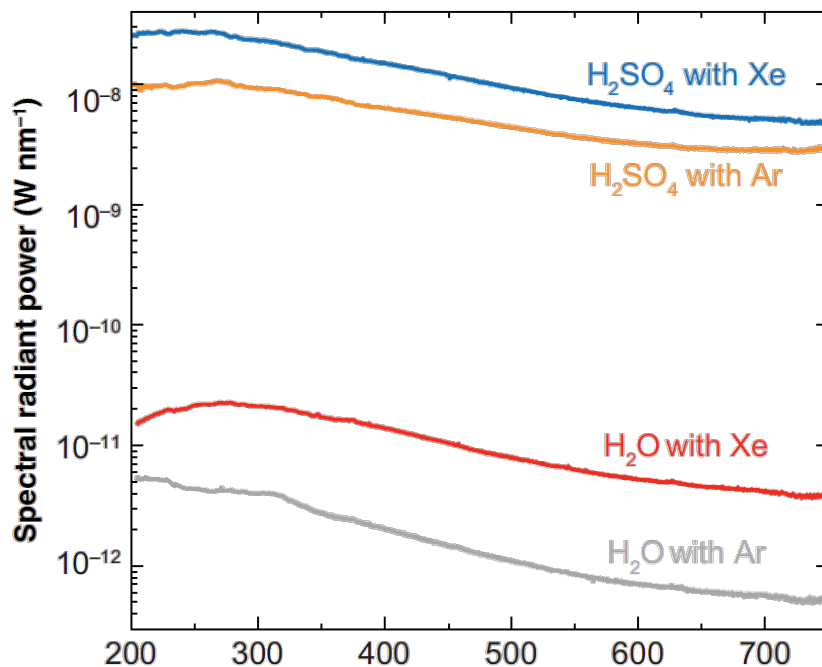
Spectra from argon and xenon bubbles in both water and 85 wt%  $\text{H}_2\text{SO}_4$



Spectrum from neon bubble in 80 wt%  $\text{H}_2\text{SO}_4$

# Ionization Energy

- Ionization energy increases as atomic number decreases
- More of the bubble is light emitting when bubble consists of lower ionization atoms



2	<b>He</b> Helium 4.00260
10	<b>Ne</b> Neon 20.1797
18	<b>Ar</b> Argon 39.948
36	<b>Kr</b> Krypton 83.80
54	<b>Xe</b> Xenon 131.29

↑ Ionization Energy

# Noble Gases and Sonoluminescence

- Aside from radiant power increasing for larger noble gas bubbles, pulse emission also increases
- Air contains 1.28 wt% argon

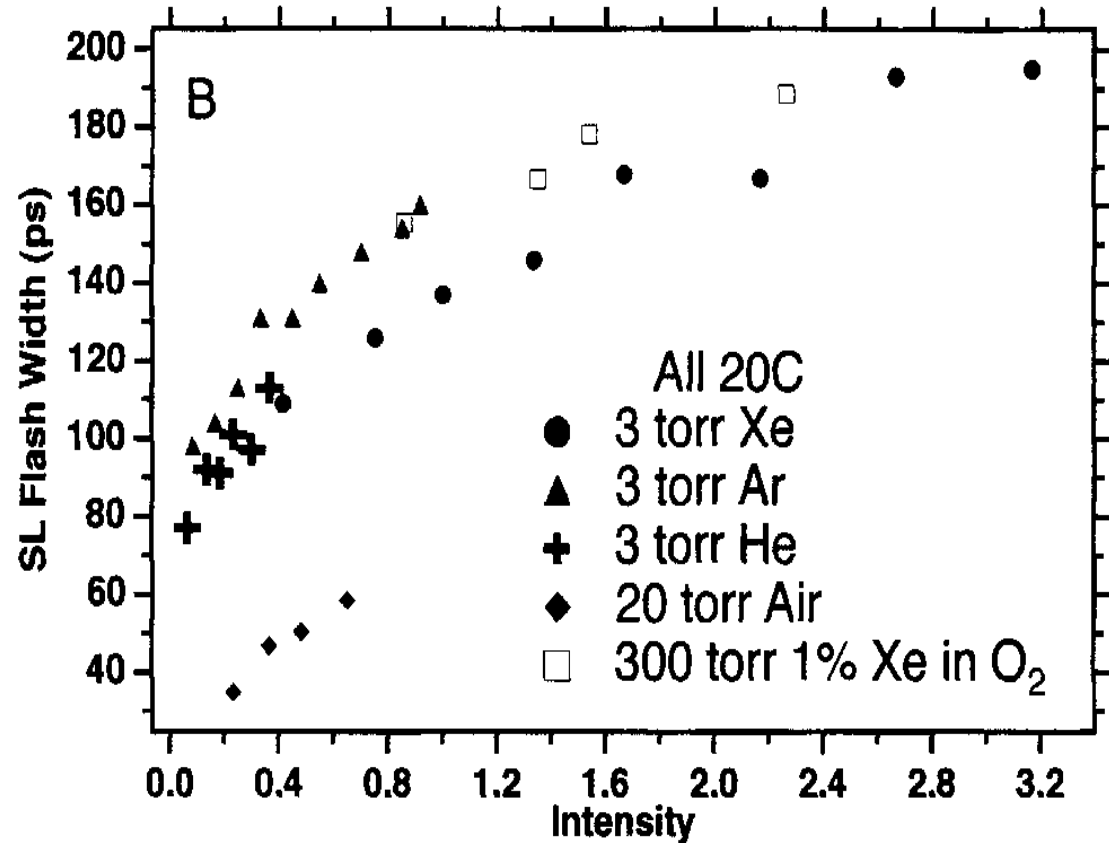
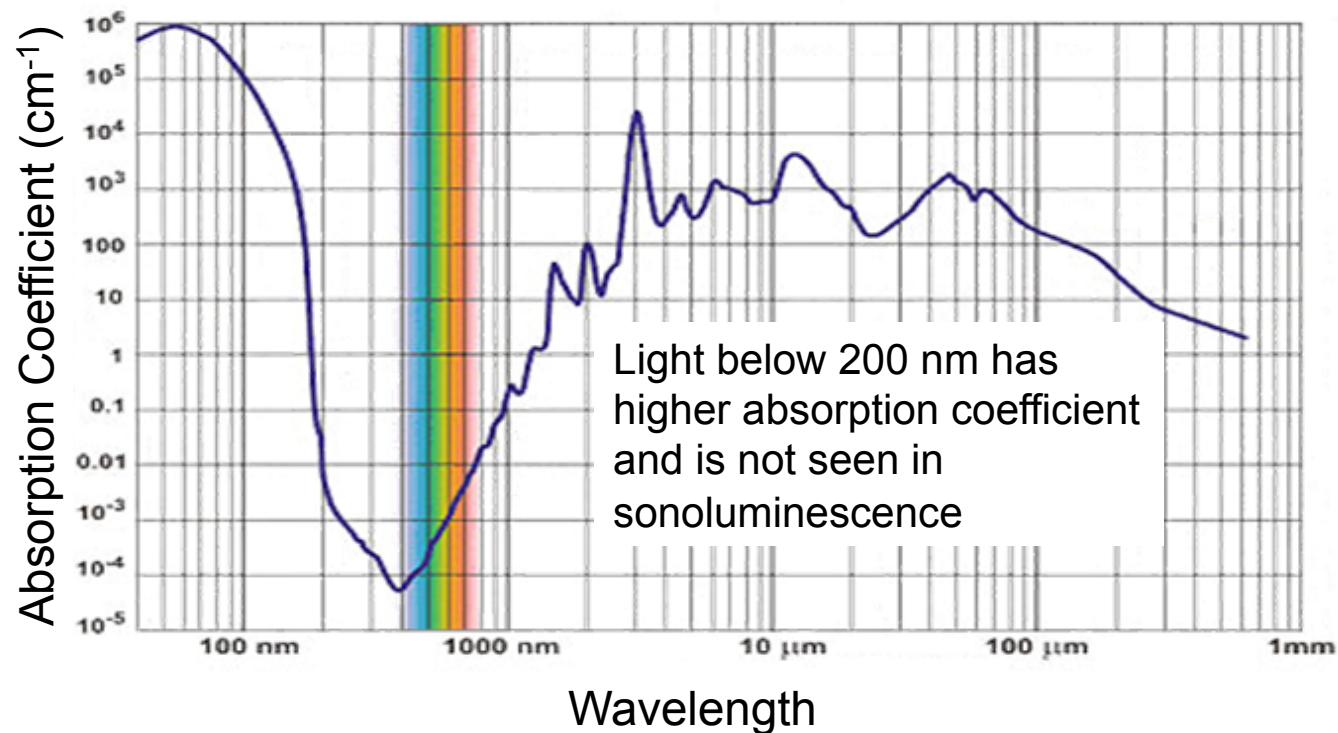


Diagram showing sonoluminescence dependence on noble gas



# Sonoluminescence Spectrum - Water absorption

- Absorption coefficient of water keeps spectrum of sonoluminescence from appearing lower than 200 nm
- Sonoluminescence wavelength window is 200-800 nm



# When is light emitted?

- Majority of light emission occurs at end of bubble collapse

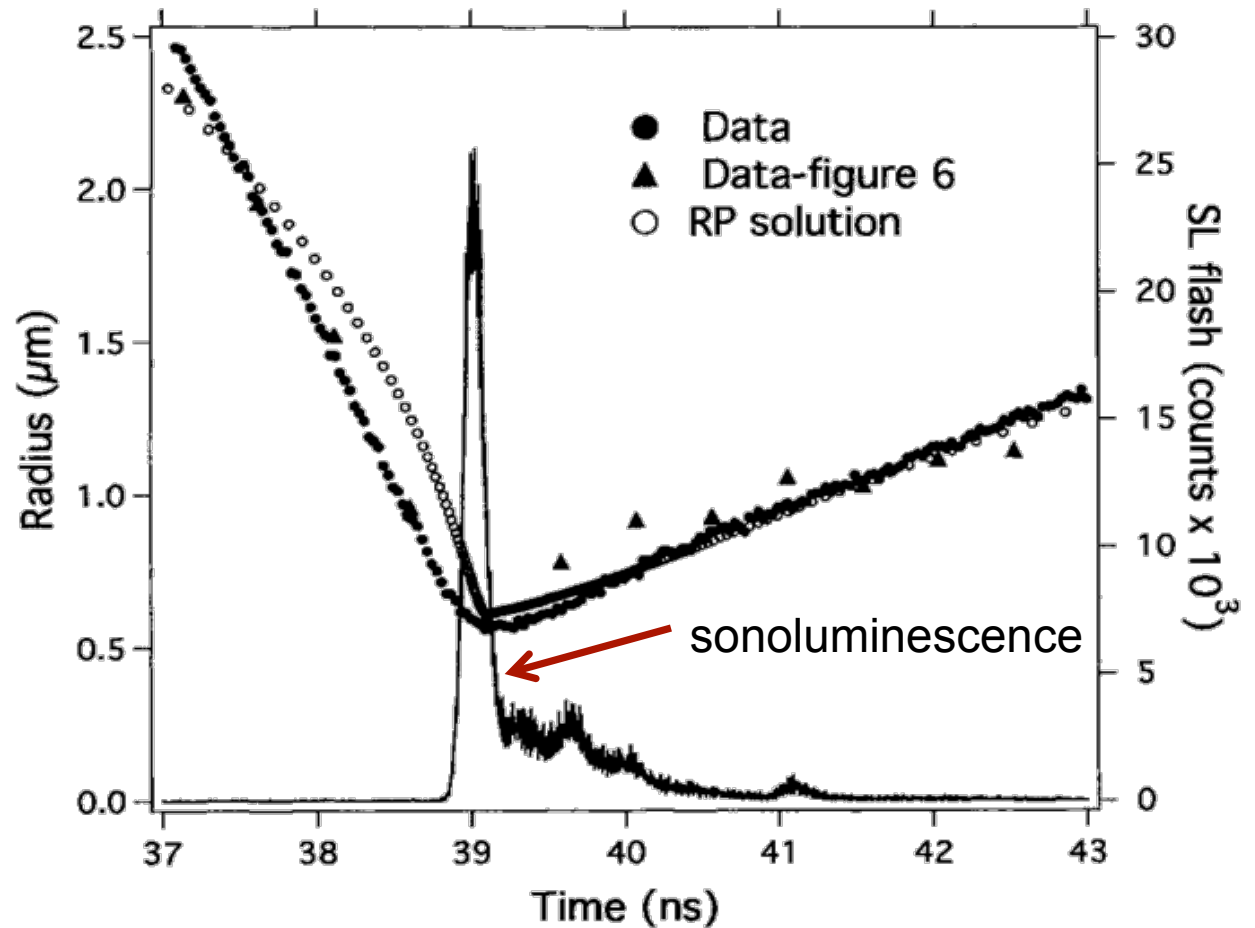


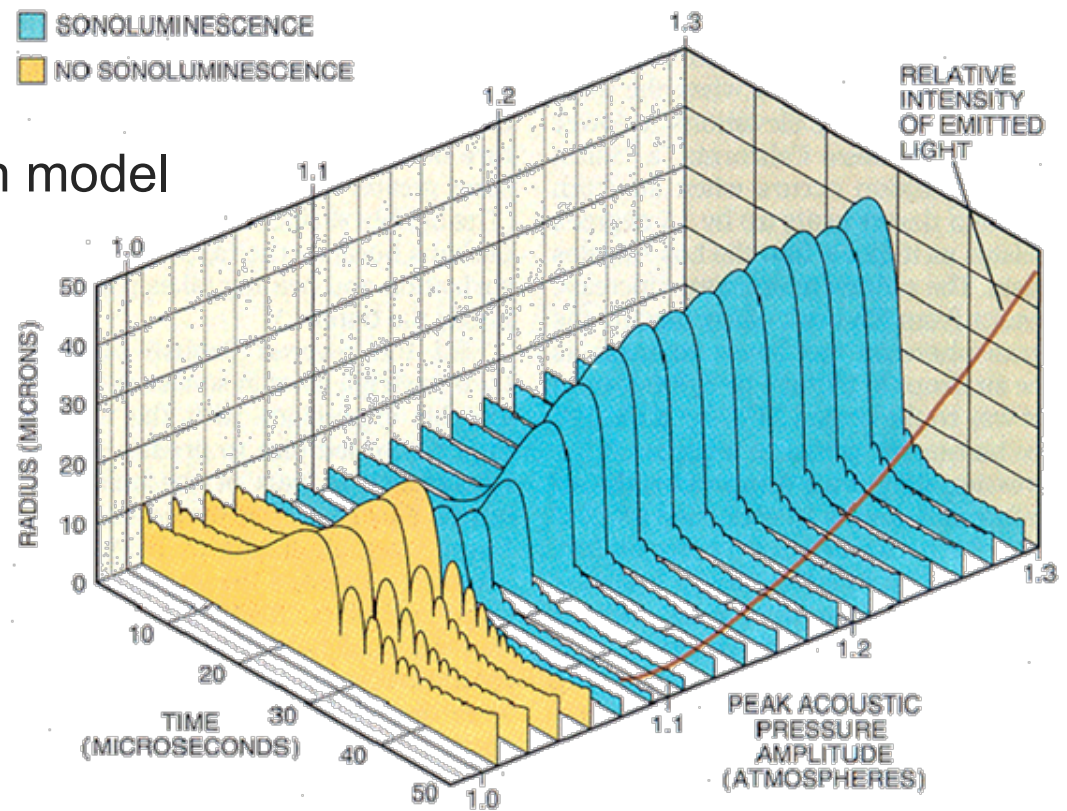
Diagram of light emission and bubble radius

Diagram of radius and light flash versus time of bubble with 1% xenon and oxygen

# What Causes Sonoluminescence?

- Theories for Cause of Sonoluminescence
  - Casimir Effect
  - Black-body radiation
  - Shockwave model
  - Quasi-adiabatic compression model

Diagram showing sonoluminescence dependence on acoustic pressure and how radial motion changes in sonoluminescence and non-sonoluminescence regime

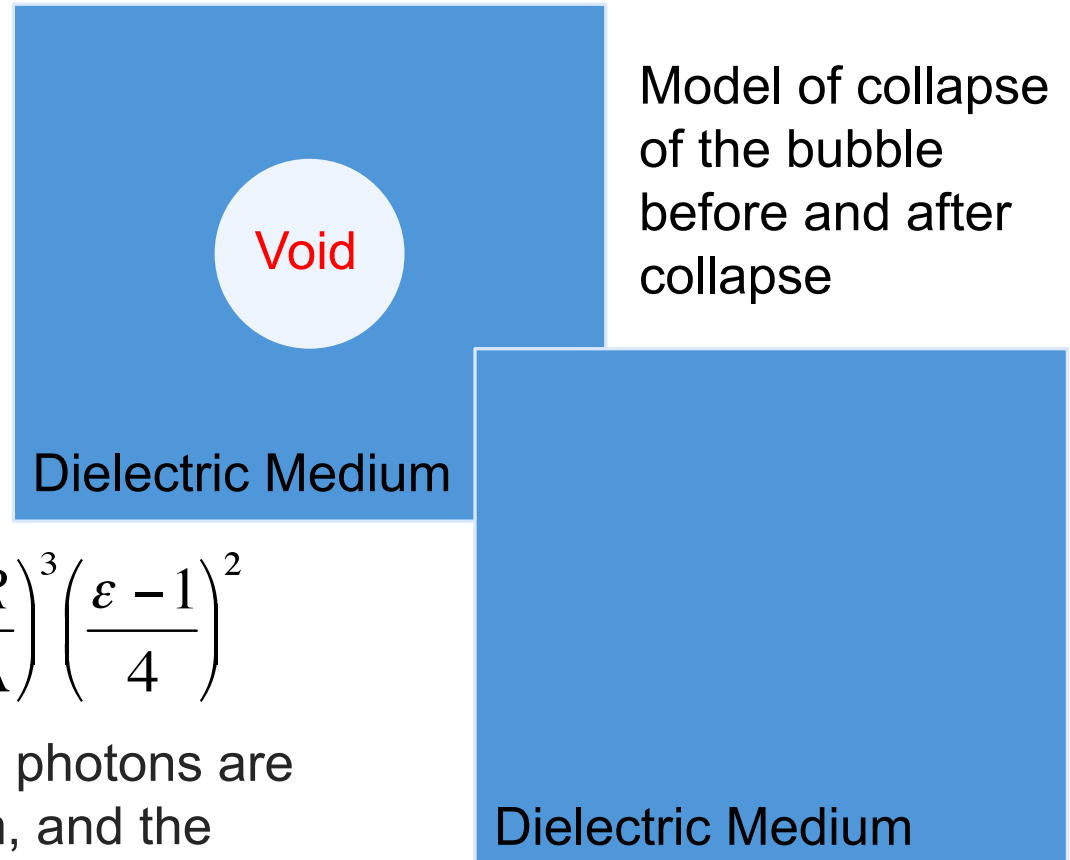


# Casimir Effect

- The Casimir effect was postulated as being a possible cause for sonoluminescence
- A simplified model would be the collapse of a void in a dielectric medium in a collapse time,  $\tau_c$

- Result of Milton, 
$$N = \left(\frac{4\pi}{3}\right)^2 \left(\frac{R}{\Lambda}\right)^3 \left(\frac{\epsilon - 1}{4}\right)^2$$

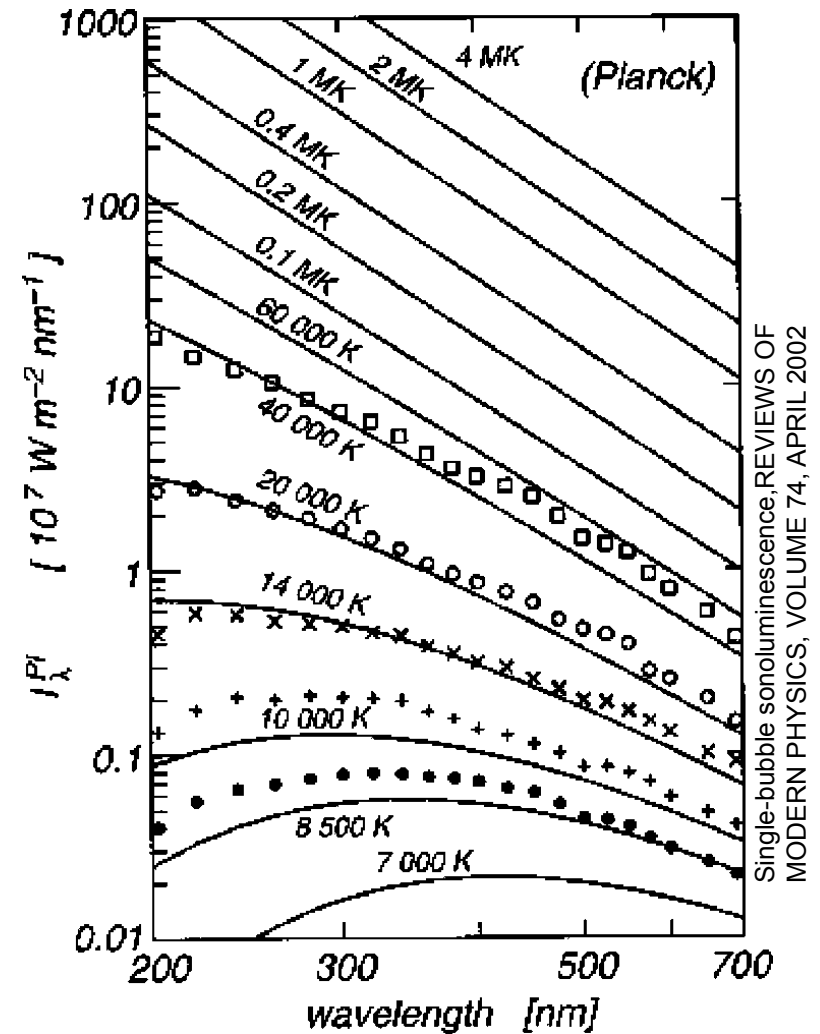
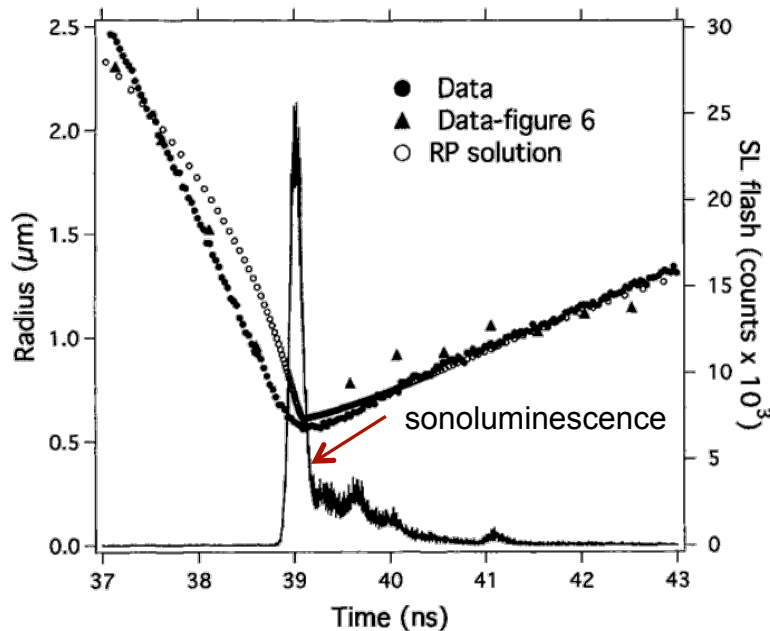
- Where R is the radius at which photons are approximately emitted, 0.5  $\mu\text{m}$ , and the cutoff wavelength  $\sim 200$  nm, the number of photons produced is  $N \sim 10$  photons five orders of magnitude too small



$N$  - photons produced  
 $\Lambda$  - cutoff wavelength  
 $\epsilon$  - relative permittivity

# Black-body Radiation

- Light would be emitted from surface
  - Bubble is a volume emitter
- During emission, bubble has larger radius for longer time
  - Bubble heats as radius decreases
  - According to model pulse width of longer wavelengths should be longer
- Red and ultraviolet pulse width are approximately equal, which contradicts black-body model



Black-body intensity for various temperatures plotted with helium, neon, argon, krypton, and xenon from top to bottom. Fit decreases as molecular weight increases

# Shockwave Model

- Center of bubble is largest source of light emission
- Center heated from shockwaves in bubble interior
- Temperature increases as shockwave reaches minimum radius
  - Heating ionizes gas at center, light emitted when gas is plasma
- Temperature decreases as shockwave expands
  - Gas ceases to be plasma
- Shockwaves not readily observed
  - Small distortions in spherical shape and low temperature gradients disrupt shockwaves

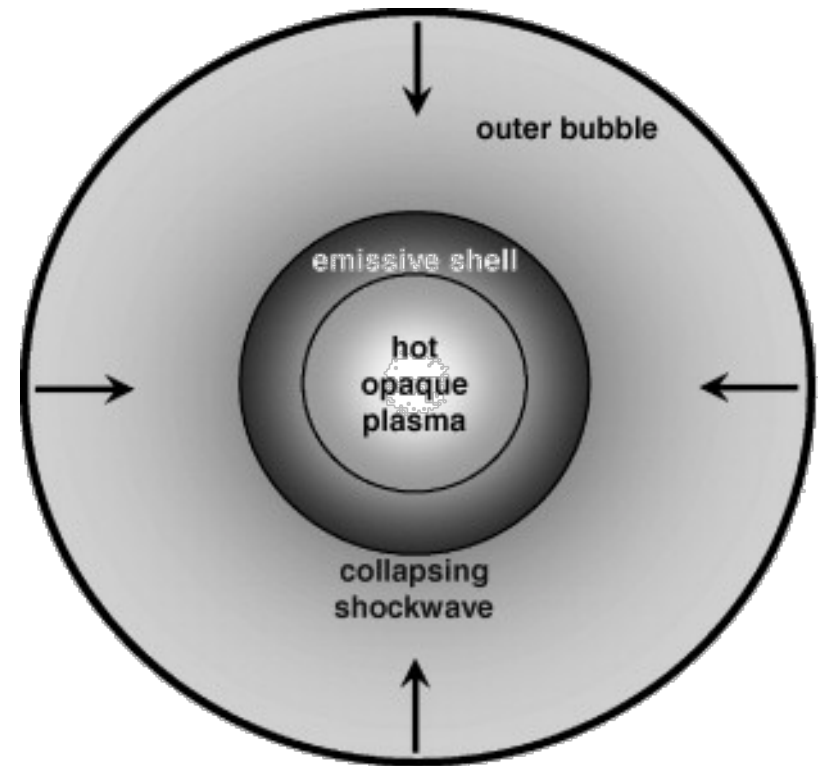
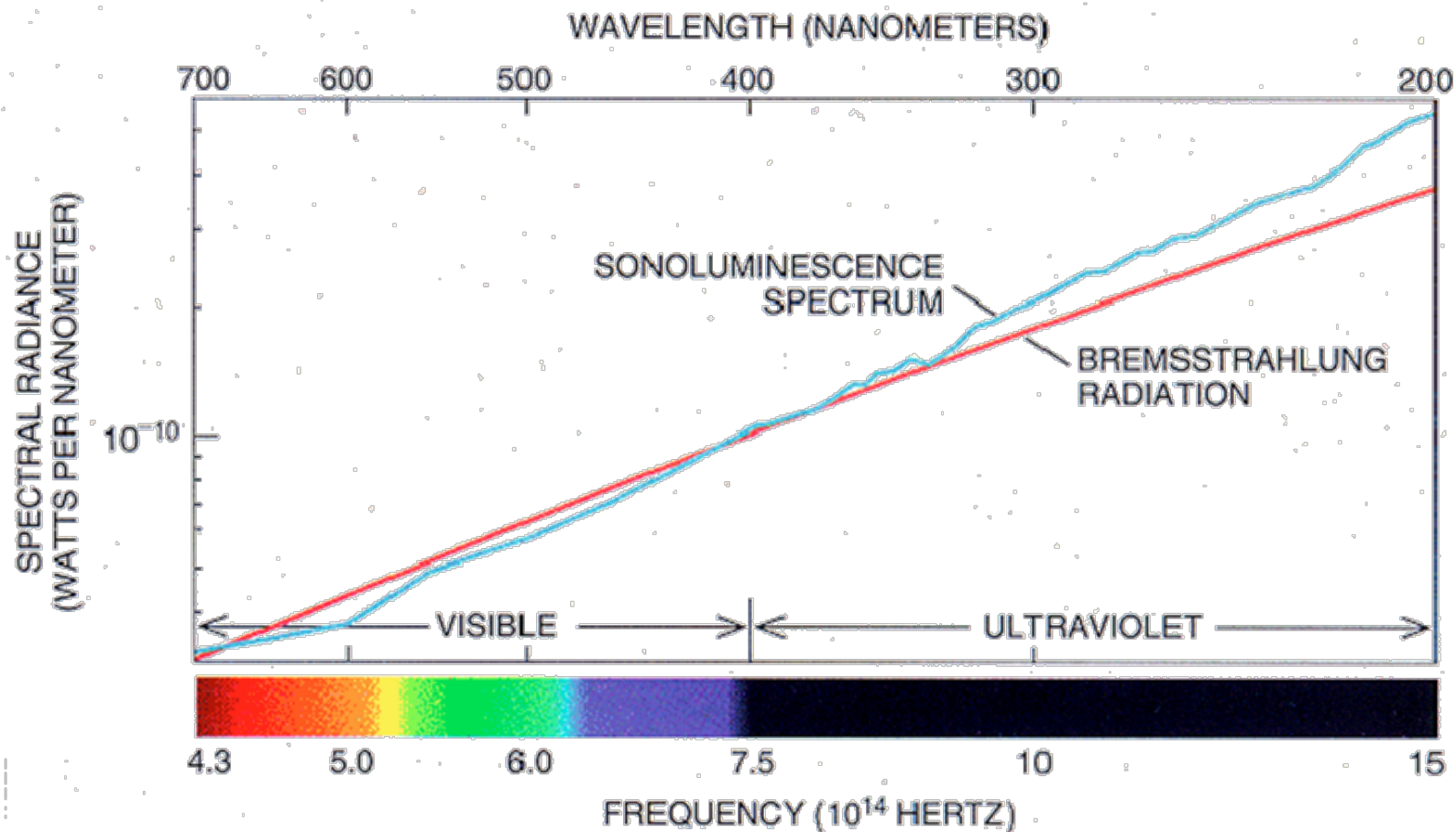


Diagram of shockwave model  
Light emitted from accelerating free electrons (Bremsstrahlung radiation)

Model fits well to spectrum provided that the shockwave reaches minimum  $\sim 0.1\mu\text{m}$

# Spectrum – Sonoluminescence and Bremsstrahlung

- This spectrum would require the plasma in the center of the bubble to reach 100,000K, requiring that the shockwave reach a radius of  $0.1\mu\text{m}$
- To reach the such a small radius, bubble asphericity would need to be minimal, as well as radiation damping and thermal diffusion



# Quasi-adiabatic compression model

- Quasi-adiabatic compression heats bubble to ionization temperatures
- Vapor at interface reduces level of ionization
- The proportions of each emission source is influenced by internal temperature of bubble
  - Electron-ion Bremsstrahlung causes less emission when surrounding temperature is hotter

Causes of Light Emission:

-Radiative attachment:  $A+e^- \rightarrow A^-+h\nu$

$$P_{att} = \langle \sigma_{att} v_e \rangle n_o n_e \frac{3}{2} kT / V$$

-Radiative recombination:  $A^+ + e^- \rightarrow A + h\nu$

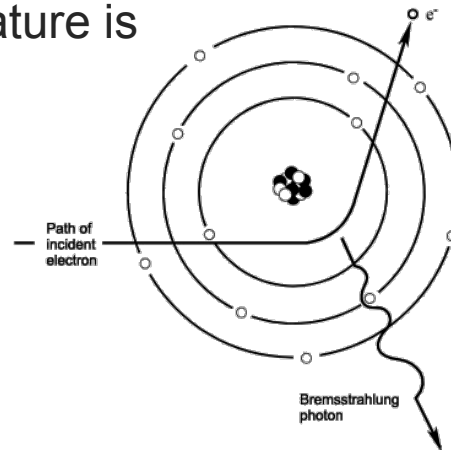
$$P_{rec} = \langle \sigma_{rec} v_e \rangle n_e^2 \frac{3}{2} kT / V \propto T^{1/2}$$

-Electron-atom Bremsstrahlung radiation

$$P_{brem,atom} = 4.6 \times 10^{-44} n_e n_{ar} T / V$$

-Electron-ion Bremsstrahlung radiation

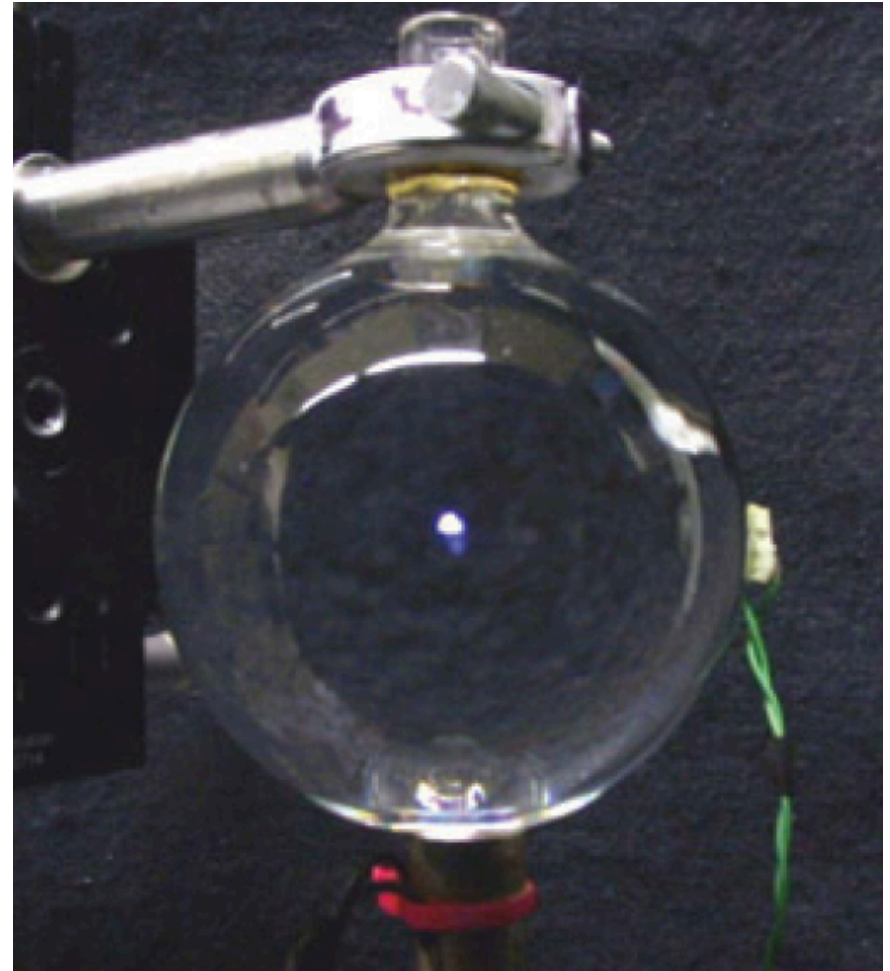
$$P_{brem,ion} = 1.57 \times 10^{-40} n_e^2 T^{1/2} / V$$





# Conclusions

- Sonoluminescence is visible light emitted from a bubble driven acoustically
- The average temperature of the bubble at collapse is  $\sim 10^3$  K
- Light is emitted in picosecond pulse widths
- Sonoluminescence is extremely sensitive to conditions
- No theory currently is agreed upon as to the cause of sonoluminescence



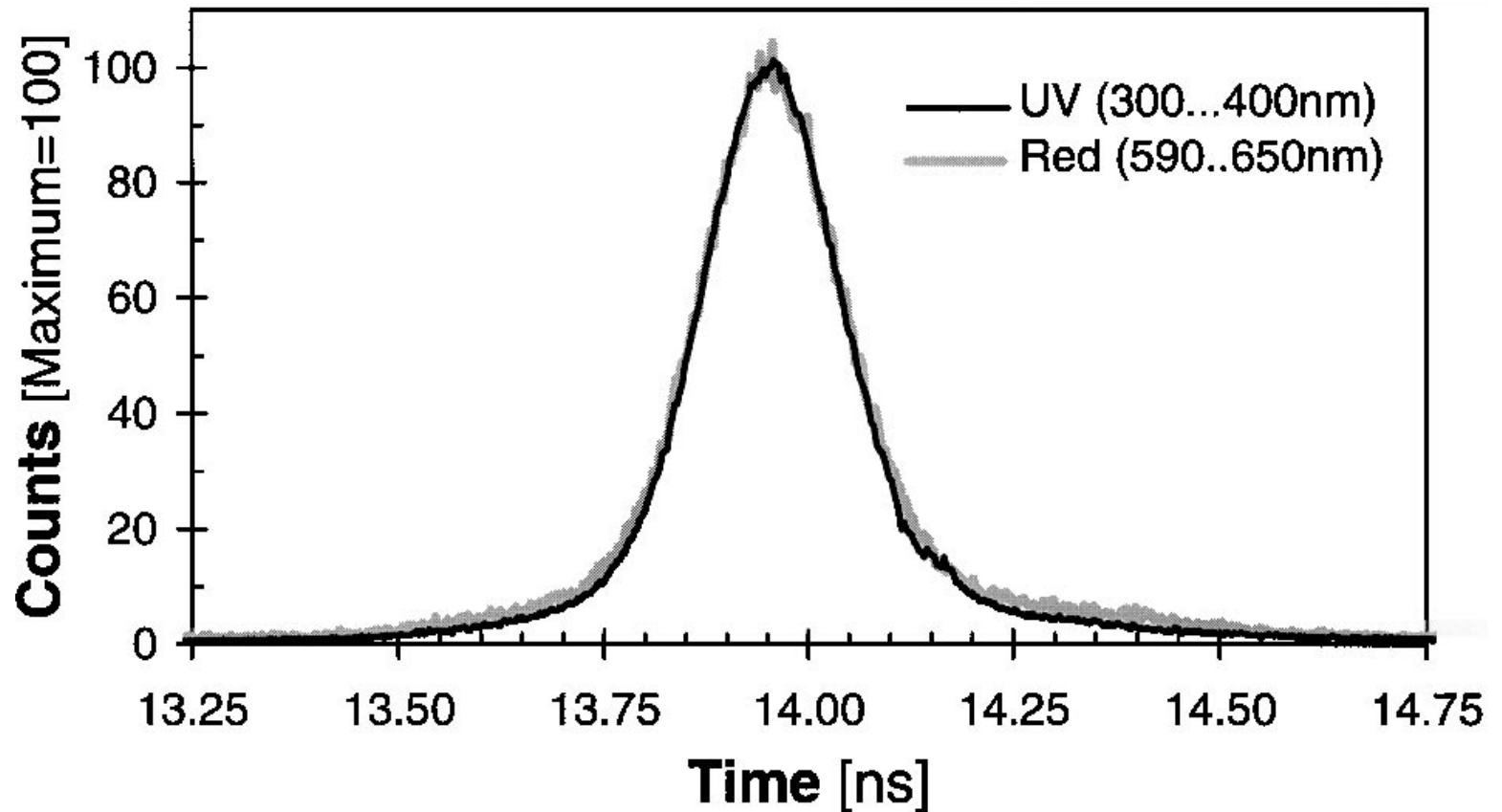
Sonoluminescent bubble of Xe in 85 wt% H<sub>2</sub>SO<sub>4</sub> in 3cm radius flask

# References

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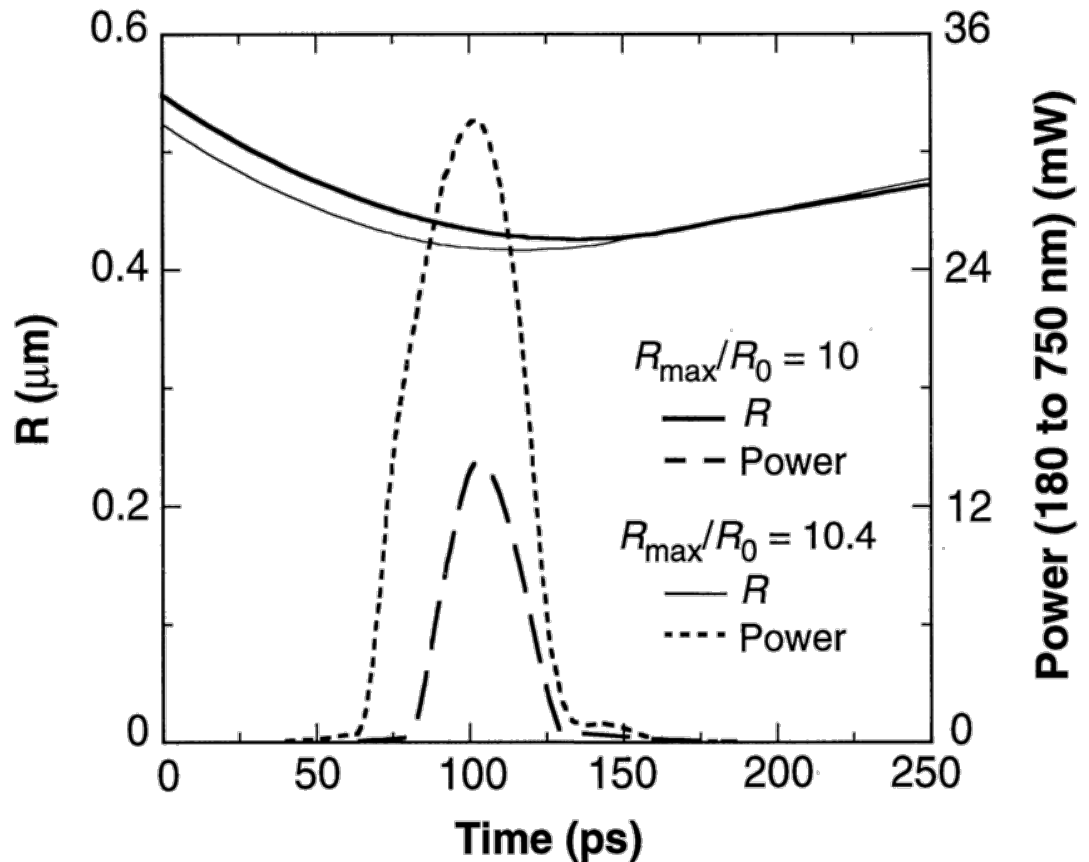
# Pulse Width for UV and Red

Red and UV parts of spectrum are nearly identical, ruling out black-body radiation



# Pressure and Sonoluminescence

- Power output is increased by factor of 2 with change in driving pressure of  $7 \cdot 10^3$  Pa
- No change in surrounding fluid temperature



Power from light emission from two bubbles with different maximum radius from difference in driving pressure of 7 kilopascal