How Do Basic Nonlinear Optical Processes Lead to Atmospheric Lasing?

(Insights from the 1980s)

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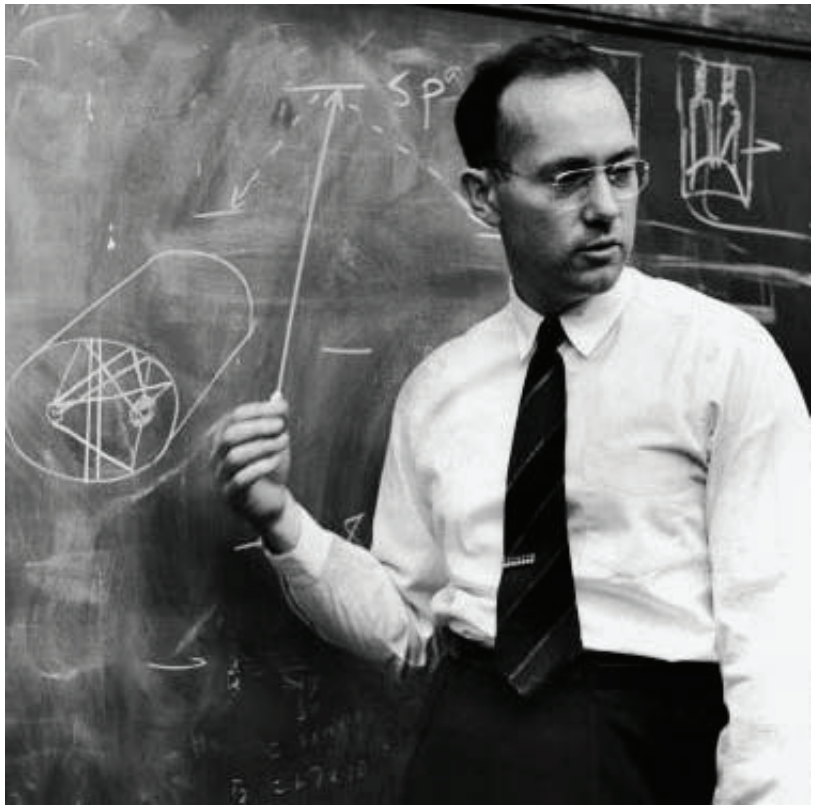
With great thanks to Paul Corkum, John Sipe, and Pavel Polynkin.

Presented at CLEO, San Jose, USA, May 11, 2015.

Charles H. Townes

July 28, 1915 to January 27, 2015



- Inventor of the maser and laser
- Nobel Prize, 1964
- Advisor to three US presidents
- Teacher, mentor, and friend

Nature 519, 292 (2015).

What is the origin of air "lasing"

Is it really "lasing," that is, operating by population inversion gain?

Some proposed mechanisms:

- mirrorless lasing (ASE)
- superfluorescence
- hyper-Raman scattering

Perhaps (probably?) each of these mechanisms can dominate under different experimental conditions. There is no one "correct" model.

Caution:

Showing that someone else's model is wrong does not make yours right!

Brief Review of Superradiance, Superfluorence, and Amplified Spontaneous Emission (ASE)

Superradiance and superfluorescence are two forms of cooperative emission.

Superfluorescence is cooperative emission from an initially totally inverted system.

Superradiance is cooperative emission from a system initially possessing a macroscopic dipole moment.

Both are characterized by a cooperative lifetime $\tau_r = \frac{8\pi A}{3\lambda^2}\frac{T_1}{N}$

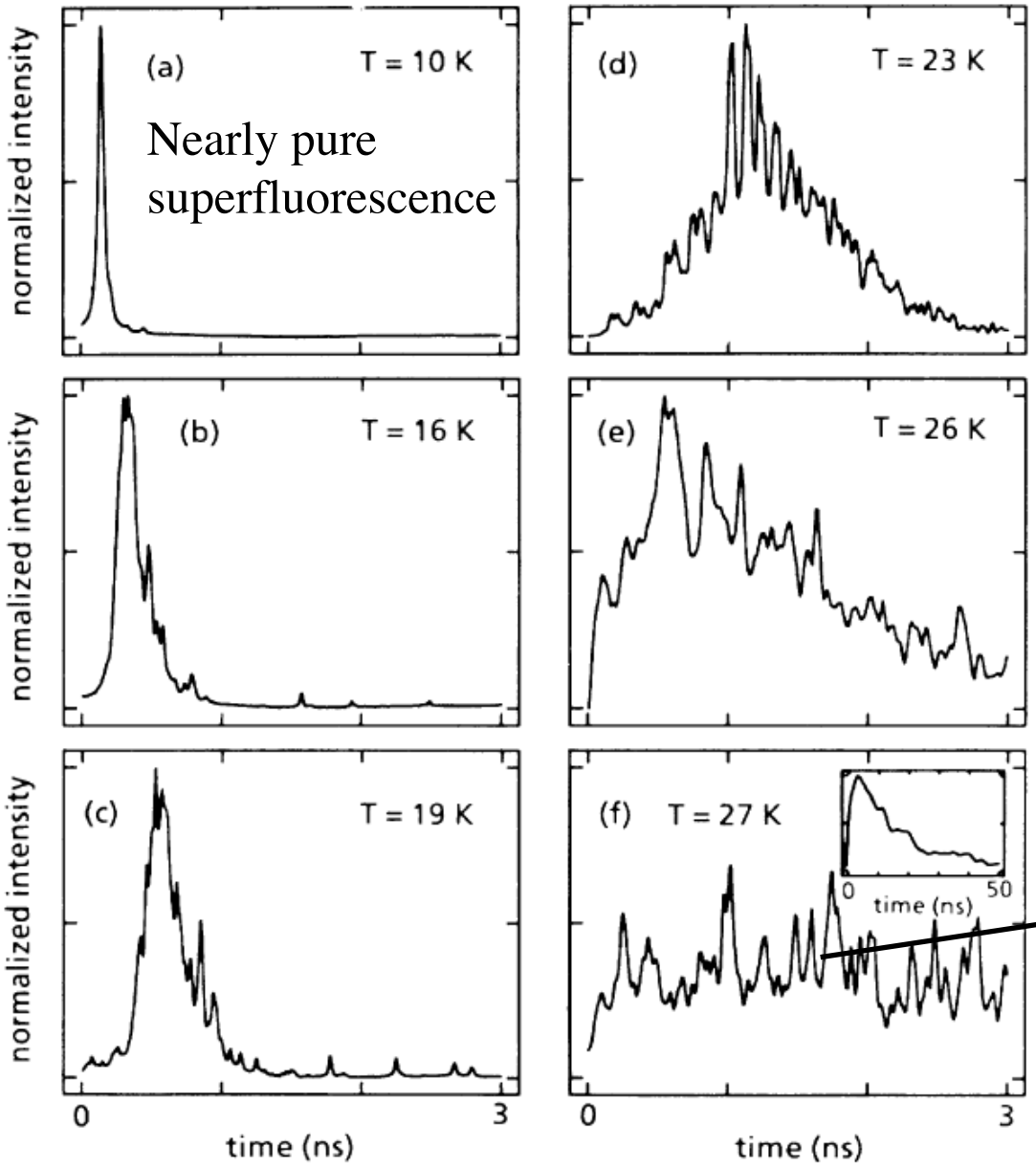
Superfluorescence is charaterized by a delay time $\tau_D = \tau_r[\frac{1}{4}\ln(2\pi N)]^2$

If the characteristic dephasing time T_2 is smaller than τ_D , a, a macroscopic dipole moment cannot develop and instead ASE occurs.

Refs: Dicke, Rehler and Eberly, Bonifacio and Lugiato, Burnham and Chiao, Gross and Haroche

Transition from Superfluorescence to Amplified Spontaneous Emission

Michelle S. Malcuit, Jeffery J. Maki, David J. Simkin,[a] and Robert W. Boyd
Institute of Optics, University of Rochester, Rochester, New York 14627
(Received 31 December 1986)

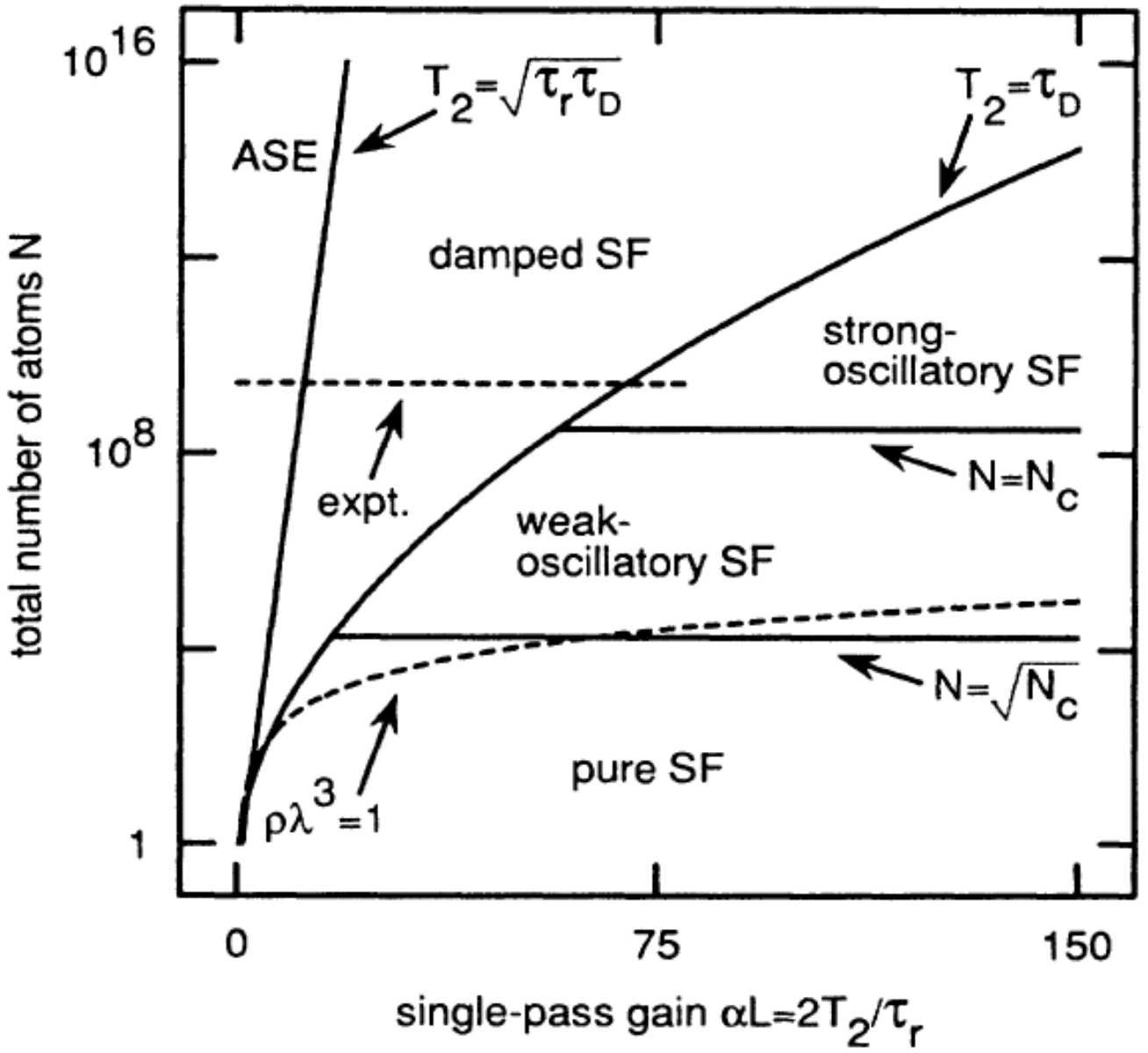


The T_2 dephasing time of $KCl:O_2^-$ is a very strong function of temperature.

By varying the temperature between 10 K and 27 K we were able to explore the transition from superfluorescence and ASE.

Note that ASE is very noisy. Not to be confused with ringing.

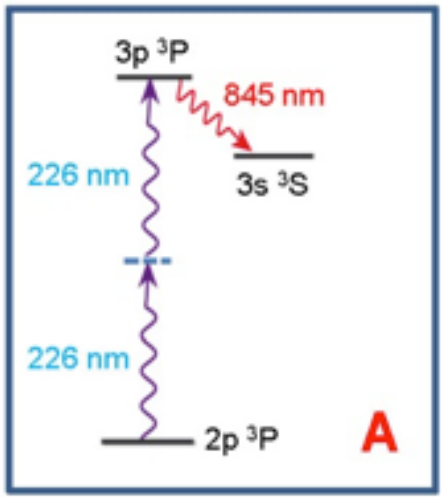
Regimes of Superfluorence

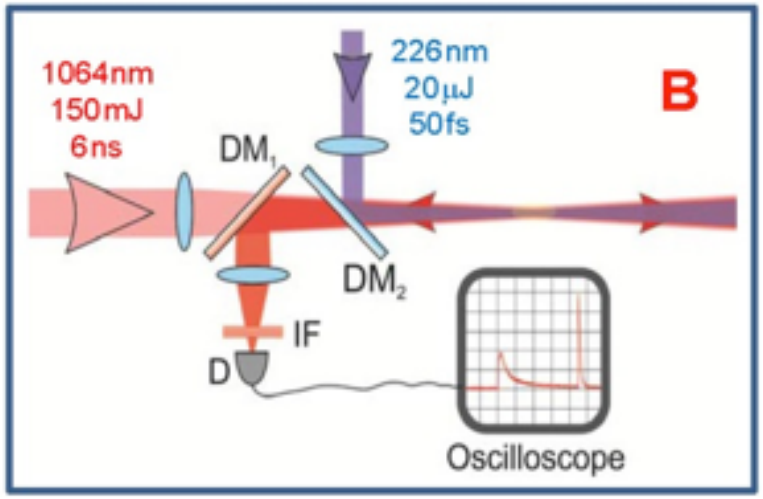


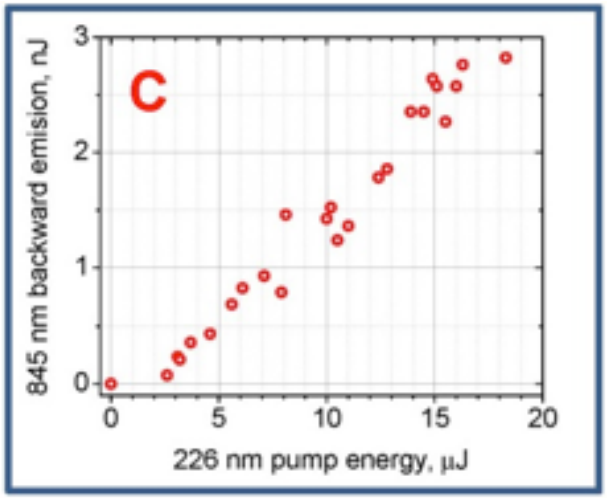
Maki, Malcuit, Raymer, Boyd and Drummond, Phys. Rev. A 40, 5135 (1989).

What is the origin of air "lasing"

A very special case: air lasing in atomic oxygen:







They find that the backward emission can be as much as 40 times stronger than the forward emission. Why is this?

Work of Alisauskas, Baltuska, and Polynkin

How to Explain Backwards Emission

The only process I know of that favors backwards emission is stimulated Brillouin scattering (SBS).

But the observation of backwards emission excited by fs pulses rules out this mechanism. (SBS has a characteristic turn-on time of approximately 1 ns.)

Processes such as ASE, stimulated Raman scattering (SRS), and stimulated hyper-Raman scattering (SHRS) have gain in both forward and backwards directions.

Is it possible that the backwards emission is due to one of these processes and that the forward emission is suppressed by some quantum interference effect?

VOLUME 55, NUMBER 10 PHYSICAL REVIEW LETTERS 2 SEPTEMBER 1985

Suppression of Amplified Spontaneous Emission by the Four-Wave Mixing Process

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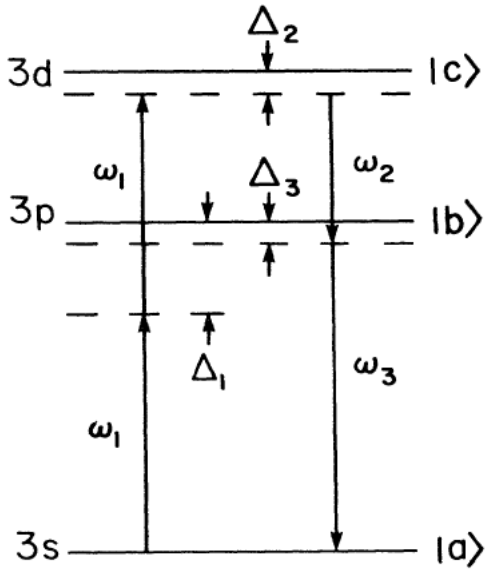
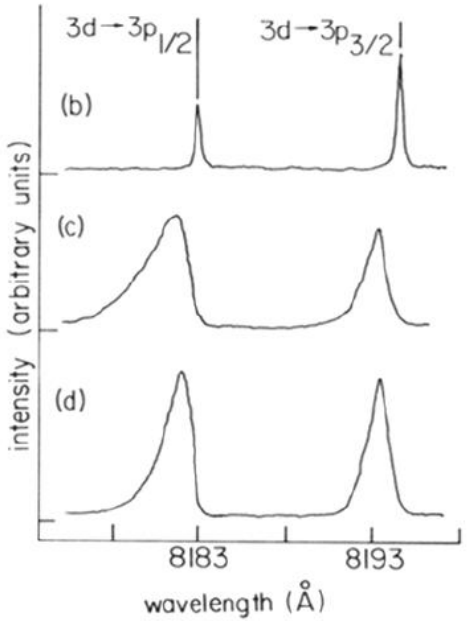
(Received 21 June



ASE can occur on the c to b transition

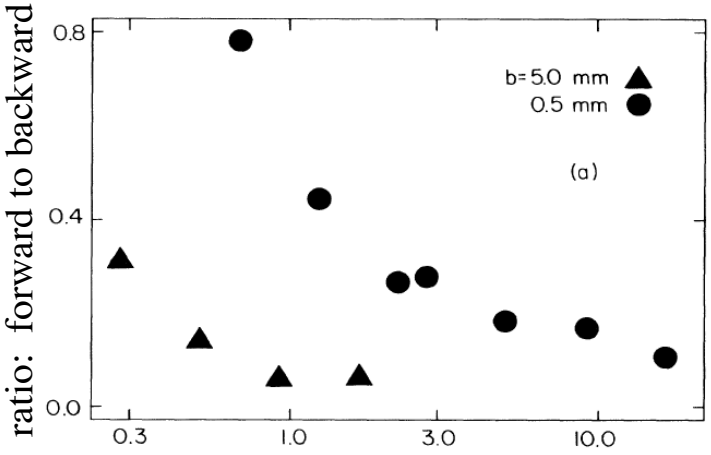


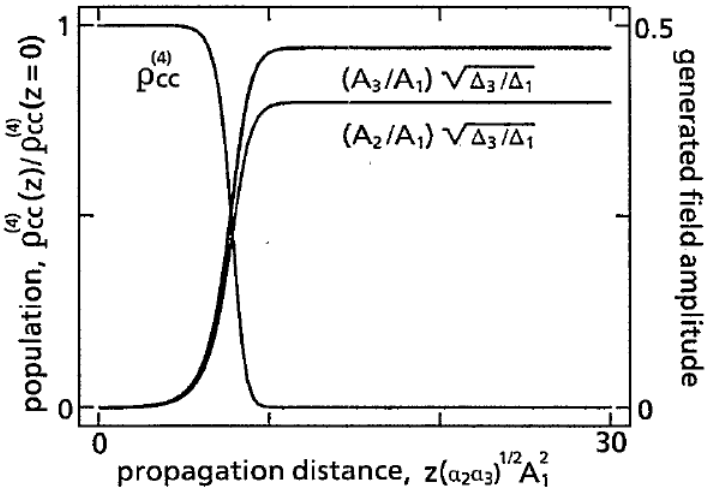
backwards (on-resonance pump) (ASE)

forward (detuned pump) (FWM)

forward (on-resonance pump) (FWM)

Forward emission suppressed at high pump intensity

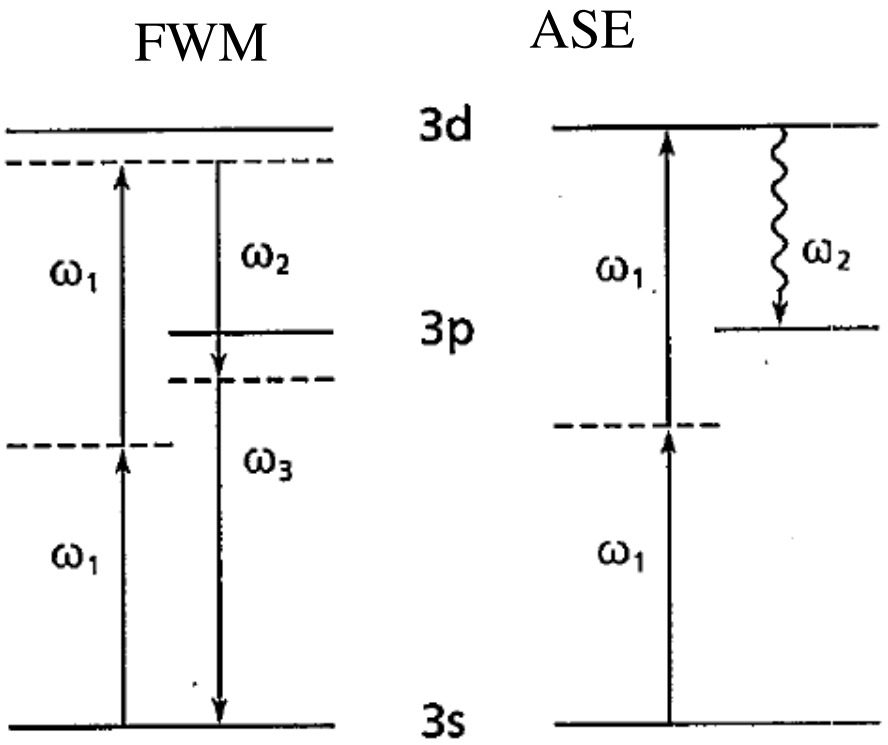




- upper-level population suppressed by FWM
- two interfering pathways to upper level

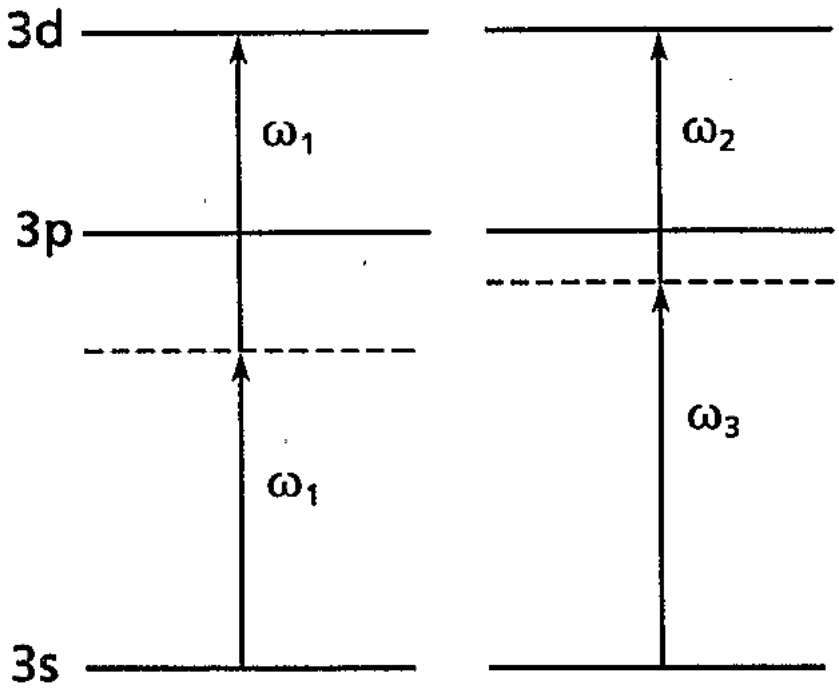
What is the Origin of the Suppression of ASE?

Both four-wave mixing (FWM) and ASE can occur



The creation of the ω_2 and ω_3 field creates a second excitation pathway to the upper level.

Under quite general conditions, these two excitation pathways interfere destructively!



Boyd, Malcuit, Gauthier, and Rzazewski, PRA 1987

What is the Origin of the Suppression of ASE?

Here are some of the details:

$$\rho_{cc}^{(4)} = -\frac{2|\mu_{ba}|^2|\mu_{cb}|^2}{\hbar^4\gamma_c}$$
$$\times \mathrm{Im}\left\{\frac{1}{\Delta_2 - i\Gamma_{ca}}\left[\frac{|\tilde{E}_1|^4}{(\Delta_1 - i\Gamma_{ba})(\Delta_2 - \Delta_1 - i\Gamma_{cb})}\right.\right.$$
$$+\tilde{E}_1^2\tilde{E}_2^*\tilde{E}_3^*\left[\frac{1}{(\Delta_1 - i\Gamma_{ba})(\Delta_2 + \Delta_3 - 2\Delta_1 - i\Gamma_{cb})} + \frac{1}{(\Delta_1 - i\Gamma_{ba})(\Delta_2 - \Delta_3 - i\Gamma_{cb})}\right]$$
$$+\tilde{E}_1^{*2}\tilde{E}_2\tilde{E}_3\left[\frac{1}{(2\Delta_1 - \Delta_3 - i\Gamma_{ba})(\Delta_2 - \Delta_1 - i\Gamma_{cb})} + \frac{1}{(\Delta_3 - i\Gamma_{ba})(\Delta_2 - \Delta_1 - i\Gamma_{cb})}\right]$$
$$+|\tilde{E}_2|^2|\tilde{E}_3|^2\left[\frac{1}{(2\Delta_1 - \Delta_3 - i\Gamma_{ba})(\Delta_2 + \Delta_3 - 2\Delta_1 - i\Gamma_{cb})}\right.$$
$$+\frac{1}{(2\Delta_1 - \Delta_3 - i\Gamma_{ba})(\Delta_2 - \Delta_3 - i\Gamma_{cb})}$$
$$\left.\left.\left.+\frac{1}{(\Delta_3 - i\Gamma_{ba})(\Delta_2 + \Delta_3 - 2\Delta_1 - i\Gamma_{cb})} + \frac{1}{(\Delta_3 - i\Gamma_{ba})(\Delta_2 - \Delta_3 - i\Gamma_{cb})}\right]\right]\right\},$$

Boyd, Malcuit, Gauthier, and Rzazewski, PRA 1987

Volume 68, number 4 OPTICS COMMUNICATIONS 15 October 1988

SUPPRESSION OF ELECTRONIC HYPER-RAMAN EMISSION BY FOUR-WAVE MIXING INTERFERENCE

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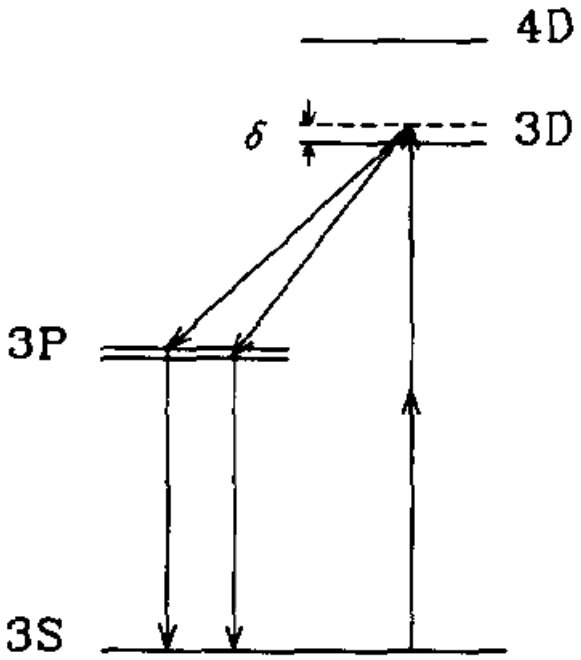
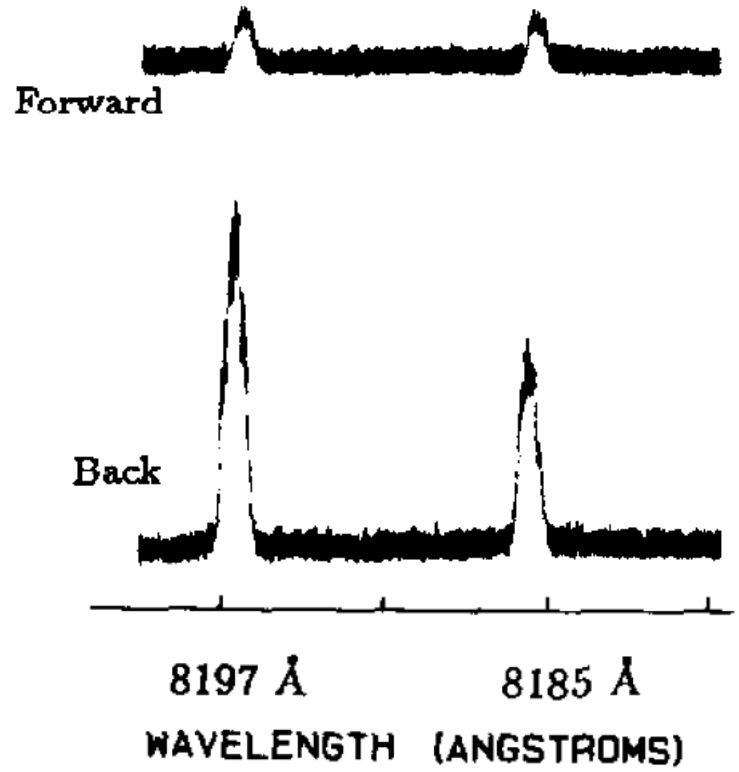
Chemical Physics Section, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6378, USA

Once again, a four-wave mixing process generates two new fields that create an additional excitation pathway for the upper (3D) level.



Observed wavelengths correspond to ASE



Discussion

I propose (along with Pavel Polynkin) that the observed backwards 845 nm emission in atomic oxygen is a created by stimulated hyper Raman scattering (SHRS).

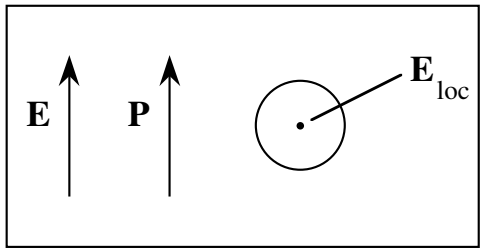
The absence of emission in the forward direction is a consequence of a destructive interference between the SHRS process and FWM.

Bonus Topic:

Local Field Effects and the Lorentz Red Shift

The Lorentz Red Shift

We recall the Lorentz-Lorenz Law of linear optics:



$$\chi = \frac{N\alpha}{1 - \frac{1}{3}N\alpha} \quad \text{or} \quad \frac{\epsilon - 1}{\epsilon + 2} = \frac{1}{3}N\alpha.$$

This result follows from the assumption that the field that acts on a representative atom is not the macroscopic Maxwell field but rather the Lorentz local field given by

$$E_{\text{loc}} = E + \frac{1}{3\epsilon_0}P \qquad \text{where} \qquad P = \epsilon_0 \chi E$$

Derivation of the Lorentz Red Shift

We introduce the standard form for the resonant contribution to the atomic polarizability

$$\alpha(\omega) = \frac{(fe^2/2m\epsilon_0\omega_0)}{\omega_0 - \omega - i\gamma}$$

(f is the oscillator strength) into the Lorentz-Lorenz law in the form

$$\frac{\epsilon - 1}{\epsilon + 2} = \tfrac{1}{3}N\alpha.$$

and solve the resulting equation for ϵ. We find that

$$\epsilon(\omega) = 1 + \frac{N(fe^2/2m\epsilon_0\omega_0)}{\omega_0 + \Delta\omega - \omega - i\gamma}$$

where

$$\Delta\omega = -N(fe^2/2m\epsilon_0\omega_0)$$

is known as the Lorentz red shift.

Discussion of the Lorentz Red Shift

The Lorentz red shift

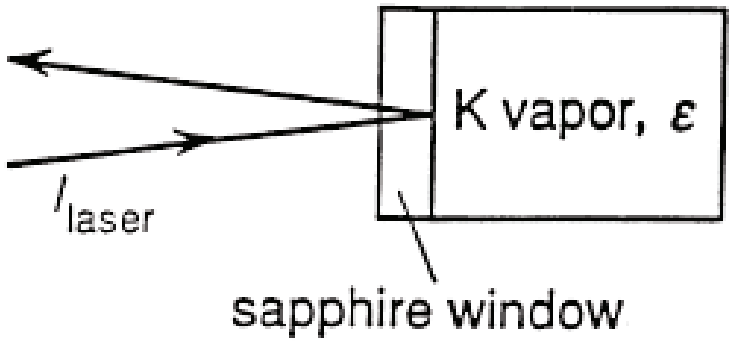
$$\Delta\omega = -N(fe^2/2m\epsilon_0\omega_0)$$

shows that the resonance frequency of the susceptibility, a macroscopic quantity, is different from that of the polarizability, a microscopic quantity.

This red-shift is purely a consequence of local-field effects, and can alternatively be understood as a sort of Lamb shift.

This result was known to Lorentz as early as 1915, but had not been verified experimentally until 1991 (Maki, Malcuit, Sipe and Boyd, PRL).

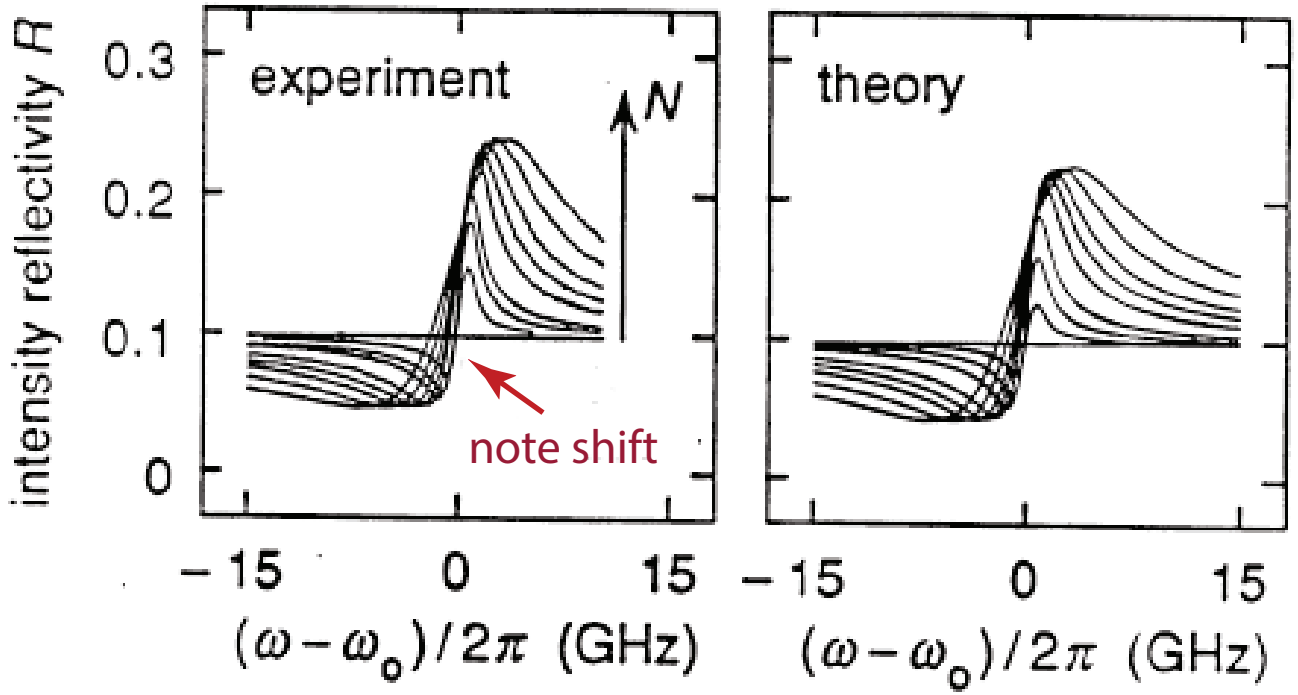
Observation of the Lorentz Red Shift



$N \sim 10^{17}\,\mathrm{cm}^{-3}$

$\Gamma_{Dop} \sim 1\,\mathrm{GHz}$

$|\varepsilon - 1| \sim 1$ and $|L| \sim 4/3$



$4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$

Maki, Malcuit, Sipe, and Boyd, Phys. Rev. Lett. 68, 972 (1991).

Thank you for your attention!

Thank you for your attention!