Lecture 23

Stimulated Raman scattering, Stokes and Anti-Stokes waves.

Raman scattering – inelastic scattering of light caused by the vibrational properties of matter

A story how energy **dissipation** can lead to field **amplification**.

After R. A. "Bob" Fisher

Raman scattering works because...

The polarizability of a *molecule* is greater than the sum of the polarizabilities of the *individual atoms* which make up the molecule!

Explanation



Atom "A"



Atom "B"

Atoms "A" and "B" are in each other's near-field. Atom A sees a field $E_{tot} = E_{ext} + E_B(A)$. Atom B sees a field $E_{tot} = E_{ext} + E_A(B)$. Here $\alpha_A(B)$ is the field measured at B which has been radiated by atom A.

The total polarization of the molecule is:

 $P_{MOLECULE} = \alpha_A E_A(tot) + \alpha_B E_B(tot).$

Here α_A is the atomic polarizability of atom A, and α_B is the atomic polarizability of atom B.

We now substitute in the two expressions for $E_B(tot)$ and $E_A(tot)$ from two vuegraphs ago to find:

 $P_{MOLECULE} = \alpha_A \{ E_{EXT} + E_B(A) \} + \alpha_B \{ E_{EXT} + E_A(B) \}$

 $= \{\alpha_A + \alpha_B\} E_{EXT} + \{\alpha_A E_B(A) + \alpha_B E_A(B)\}$

extra term





 $E_A(B)$ and $E_B(A)$ are both functions of the distance between atoms A and B.

Since the molecule vibrates at frequency Ω , the distance between A and B oscillates sinusoidally.

Therefore the polarizability of the molecule is oscillating periodically in time with frequency Ω



Forward action

Molecule: vibrates at freq. $\boldsymbol{\Omega}$



sidebands $\pm \Omega$ are created



This modulated intensity coherently excites the molecular oscillation at frequency

 $\omega_{\rm L}$ – $\omega_{\rm S}$ = Ω .



Two laser frequencies force molecule to vibrate at freq. Ω



 $F = \frac{dW}{dq} = \frac{1}{2} \frac{d(\epsilon_0 \alpha E^2)}{dq} = \frac{1}{2} \epsilon_0 \left(\frac{\partial \alpha}{\partial q}\right) E^2$

total field $\mathcal{E}(t) = \frac{1}{2} (E_L e^{i(\omega_L t - k_L z)} + E_S e^{i(\omega_S t - k_S z)} + c.c.)$

The force term oscilalting at Ω is related to E^2 : $\rightarrow \frac{1}{4}(E_L e^{i(\omega_L t - k_L z)} + E_S e^{i(\omega_S t - k_S z)} + c.c.)^2$

$$F(z,t) = \frac{1}{2}\epsilon_0 \left(\frac{\partial\alpha}{\partial q}\right) \times 2 \times \frac{1}{4} \left(E_L E_S^* e^{i(\Omega t - Kz)} + c.c.\right) = \frac{1}{4}\epsilon_0 \left(\frac{\partial\alpha}{\partial q}\right) \left(E_L E_S^* e^{i(\Omega t - Kz)} + c.c.\right)$$

$$K = k_L - k_S \text{ and } \Omega = \omega_L - \omega_S.$$

Simple oscillator model for molecular motion.

$$\ddot{q} + \gamma \dot{q} + \Omega_0^2 q = \frac{F(t)}{m} = \frac{1}{m} \frac{1}{4} \epsilon_0 \left(\frac{\partial \alpha}{\partial q}\right) \left(E_L E_S^* e^{i(\Omega t - Kz)} + c.c.\right)$$
look for q in the form $q = \frac{1}{2} [q(\Omega) e^{i(\Omega t - Kz)} + c.c.]$

we thus find that the amplitude of the molecular vibration is given by

$$q(\Omega) = \frac{1}{2m} \epsilon_0 \left(\frac{\partial \alpha}{\partial q}\right) \frac{E_L E_S^*}{\Omega_0^2 - \Omega^2 + i\omega\gamma}$$

NL polarization $P(z,t) = Np(z,t) = N\epsilon_0 \alpha(z,t)E(z,t) = \epsilon_0 N(\alpha_0 + \left(\frac{\partial \alpha}{\partial q}\right)q)E(z,t)$

the nonlinear part of the polarization oscillating at Ω is given by

$$P(z,t)_{NL} = \epsilon_0 N \left(\frac{\partial \alpha}{\partial q}\right) \frac{1}{2} (q(\Omega)e^{i(\Omega t - Kz)} + c.c.) \frac{1}{2} (E_L e^{i(\omega_L t - k_L z)} + E_S e^{i(\omega_S t - k_S z)} + c.c.)$$

The part of this expression that oscillates at frequency ω_S – the Stokes polarization is given by

Fourier component of NL polariz.

$$P(\omega_{S}) = \frac{1}{2}\epsilon_{0}N\left(\frac{\partial\alpha}{\partial q}\right)q(\Omega)^{*}E_{L} = \frac{1}{2}\epsilon_{0}N\left(\frac{\partial\alpha}{\partial q}\right)\frac{1}{2m}\epsilon_{0}\left(\frac{\partial\alpha}{\partial q}\right)\frac{E_{L}E_{L}^{*}E_{S}}{\Omega_{0}^{2} - \Omega^{2} - i\omega\gamma} = \frac{\epsilon_{0}^{2}N}{4m}\left(\frac{\partial\alpha}{\partial q}\right)^{2}\frac{|E_{L}|^{2}E_{S}}{\Omega_{0}^{2} - \Omega^{2} - i\omega\gamma}$$

Now plug Fourier component into SVEA equation:
$$\frac{\partial E(\omega_i)}{\partial z} = -\frac{i\omega_i}{2nc\epsilon_0} P_{NL}(\omega_i)$$

to get:

$$\frac{dE(\omega_S)}{dz} = -\frac{i\omega_S}{2nc\epsilon_0} \frac{\epsilon_0^2 N}{4m} \left(\frac{\partial\alpha}{\partial q}\right)^2 \frac{|E_L|^2 E(\omega_S)}{\Omega_0^2 - \Omega^2 - i\omega\gamma} e^{-\Delta kz}$$

Note that $\Delta k=0$ here – SRS is always phase matched, since

$$\Delta k = k_L - k_L + k_S - k_S = 0 (23.1)$$

Thus Stokes wave can propagate in any direction, even counter-propagate!

Let us now set $\Omega = \Omega_0$

$$\frac{dE(\omega_{S})}{dz} = -\frac{i\omega_{S}}{8nc\epsilon_{0}} \frac{\epsilon_{0}^{2}N}{m} \left(\frac{\partial\alpha}{\partial q}\right)^{2} \frac{|E_{L}|^{2}E(\omega_{S})}{-i\omega\gamma} = \frac{1}{8nc\epsilon_{0}} \frac{\epsilon_{0}^{2}N}{m\gamma} \left(\frac{\partial\alpha}{\partial q}\right)^{2} |E_{L}|^{2}E(\omega_{S}) = \alpha E(\omega_{S})$$

At exact resonance, purely real

exponential growth

$$\frac{dE(\omega_S)}{dz} = \alpha E(\omega_S) \qquad (23.2)$$

$$\alpha = \frac{1}{8nc\epsilon_0} \frac{\epsilon_0^2 N}{m\gamma} \left(\frac{\partial \alpha}{\partial q}\right)^2 |E_L|^2 = \frac{1}{4} \frac{\epsilon_0^2 N}{m\gamma} \left(\frac{\partial \alpha}{\partial q}\right)^2 I_L$$



 $\omega_L - \omega_S$ detuning from resonance

How wide is the SRS gain bandwidth ? - from GHz to many THz

Raman frequency shift: 15.6 THz in silicon 120 THz in H₂ gas

Quantum description

Nonlinear susceptibility $\chi^{(3)}$, quantum mechanical model

According to Boyd, Stegeman

 $\chi^{(3)}$ - is a huge sum of many different terms



$$\chi_{kjih}^{(3)}(\omega_{\sigma},\omega_{r},\omega_{q},\omega_{p}) = \frac{N}{\epsilon_{0}\hbar^{3}} \mathcal{P}_{F} \sum_{mn\nu} \frac{\mu_{g\nu}^{k} \mu_{\nu n}^{j} \mu_{nm}^{i} \mu_{mg}^{h}}{(\omega_{\nu g} - \omega_{\sigma})(\omega_{ng} - \omega_{q} - \omega_{p})(\omega_{mg} - \omega_{p})},$$

where $\omega_{\sigma} = \omega_p + \omega_q + \omega_r$ and where we have made use of the full permutation operator \mathcal{P}_F defined following Eq. (3.2.28).

- here frequenices $\omega_p \ \omega_q \ \omega_r$ can be both positive and negative and can each take values of $\pm \omega_1 \pm \omega_2 \pm \omega_3 \pm \omega_4$

Let us assume we have only two frequencies ('Laser' and 'Stokes') ω_L and ω_S ($\omega_S < \omega_L$) and take the terms that are close to the **Raman** resonance, that is ω_{21} is close to $\omega_L - \omega_S$

$$\chi^{(3)} \approx \frac{N}{\epsilon_0 \hbar^3} \mu_{14} \mu_{12} \mu_{23} \mu_{34}$$

$$\times \sum_{p,q,r} \left[\frac{1}{[\omega_{21} - (\omega_L - \omega_S)](\omega_{41} - \omega_p - \omega_q - \omega_r)(\omega_{31} - \omega_p)} + \ldots \right]$$



resonant term - what is in brackets is close to zero



damping term added

 \rightarrow



total NL polariz.
$$\mathcal{P}_{NL}(t) = \frac{1}{8} \epsilon_0 \chi^{(3)} (E_L e^{i\omega_L t} + E_S e^{i\omega_S t} + c.c.)^3$$

Now let us pick only components with $\pm \omega_S$ - due to interaction of three waves

$$\mathcal{P}_{NL}(t)|_{\omega_{S}} = \frac{1}{8}\epsilon_{0}\chi^{(3)}(6E_{L}E_{L}^{*}E_{S}e^{i\omega_{S}t} + c.c.) = \frac{3}{4}\epsilon_{0}\chi^{(3)}(E_{L}E_{L}^{*}E_{S}e^{i\omega_{S}t} + c.c.)$$

Fourier component $P(\omega_S) = \frac{3}{2} \epsilon_0 \chi^{(3)} E_L E_L^* E_S$

Now plug into SVEA eqauation:

$$\frac{\partial E(\omega_S)}{\partial z} = -\frac{i\omega_S}{2nc\epsilon_0} P_{NL}(\omega_i)$$

$$\frac{dE(\omega_S)}{dz} = -\frac{i\omega_S}{2nc\epsilon_0} \frac{3}{2}\epsilon_0 \chi^{(3)} E_L E_L^* E_S e^{-\Delta kz} = -i\frac{3}{4}\frac{\omega_S \chi^{(3)}}{nc} E_L E_L^* E_S e^{-\Delta kz}$$
$$\Delta k = 0$$
$$\chi^{(3)} = \chi_R^{(3)} + i\chi_I^{(3)} - \text{real and imaginary parts}$$

If we set:

Real part - responsible for cross-phase modulation Imaginary part - responsible for **Raman gain**

$$\frac{dE(\omega_S)}{dz} = \frac{3}{4} \frac{\omega_S \chi_I^{(3)}}{nc} |E_L|^2 E(\omega_S) = \frac{3}{4} \frac{\omega_S \chi_I^{(3)}}{nc} \frac{2I_L}{\epsilon_0 nc} E(\omega_S) = \frac{3}{2} \frac{\omega_S \chi_I^{(3)}}{\epsilon_0 n^2 c^2} I_L E(\omega_S)$$

For intensity $\frac{dI_S}{dz} = g_R I_L I_S$ $I_S = I_{S0} e^{g_R I_L L}$ (23.3) $\frac{I_S}{I_{S0}} - 1$



Raman gain coeff. measured in *m/W*

After R. A. "Bob" Fisher

Stimulated Raman scattering

Input wave + *ϵ*(Raman excitation) ➡ Output Wave (mechanism is Bragg scattering)

Input wave + ϵ (output wave) \implies Raman excitation

(Mechanism is that the interference term drives the vibrations)

Stimulated Raman emission in a glass fiber

276 Appl. Phys. Lett., Vol. 22, No. 6, 15 March 1973

Raman gain in glass optical waveguides

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The small signal Raman gain in a single-mode glass waveguide amplifier has been measured directly. The measured gain is in good agreement with that calculated from the Raman cross section. The cross section was determined by a comparison of the spontaneous Raman scattering of fused quartz and benzene.



Fiber : fused quartz (3.8 µm core diam.) Pump: Xenon laser at λ =526 nm Signal: at λ =535.3 nm (Raman frequency shift 330 cm⁻¹ or 10 THz) Measured Raman gain coeff. $g = 1.5 \times 10^{-11} \text{ cm/W}$

> The Raman gain coefficient is determined from the measured amplification using

$$P_s/P_{s0} = \exp(gI_{p0}L),$$



FIG. 2. Raman gain of fused quartz plotted as a function of frequency shift from an exciting line at 526 nm. The experimental point is the gain measured in the amplifier and the error bar represents a combination of the uncertainties both in the measurement of the gain and the spontaneous cross section.

Example: for the pump power of 100W $(I_p = 0.75 \text{ GW/cm}^2)$ and L=590 cm

we get : Gain = $\exp(gI_pL) = \exp(6.6) = 740$

Substance	Frequency	Linewidth Δv	Cross Section	Gain
	Shift v_0	(cm^{-1})	$N(d\sigma/d\Omega)_0$	Factor ^b
	(cm^{-1})		$(10^{-6} \text{ m}^{-1} \text{ sec}^{-1})$	G/I_L (m/TW)
Liquid O ₂	1552	0.117	0.48 ± 0.14	145
Liquid N ₂	2326.5	0.067	0.29 ± 0.09	160 ± 50
Benzene	992	2.15	3.06	28
CS ₂	655.6	0.50	7.55	240 -> 24 cm
Nitrobenzene	1345	6.6	6.4	21 GV
Bromobenzene	1000	1.9	1.5	15
Chlorobenzene	1002	1.6	1.5	19
Toluene	1003	1.94	1.1	12
NiNbO3	256	23	381	89
	637	20	231	94
Ba2NaNb5O15	650			67
LiTaO3	201	22	238	44
SiO ₂	467			08
Methane gas	2916		(10 atm) c	6.6
H ₂ gas	4155		(>10 atm)	15
H ₂ gas (rotat.)	450		(>0.5 atm)	5
Deuterium gas	2991		(>10 atm)	11
N ₂ gas	2326		(10 atm) c	0.71
O ₂ gas	1555		(10 atm) ^c	0.16

TABLE 10.2.1 Properties of stimulated Raman scattering for several materials a^{a}

^{*a*} After Kaiser and Maier (1972) and Simon and Tittel (1994). All transitions are vibrational except for the 450 cm⁻¹ hydrogen transition which is rotational.

^b Measured at 694 nm unless stated otherwise.

^c Measured at 500 nm.

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Prospects for Silicon Mid-IR Raman Lasers
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(Invited Paper)

Solid-state Raman materials

			LIO		C WO
Properties of Raman	Silicon	$Ba(NO_3)_2$	LIIO ₃	$KGd(WO_4)_2$	CaWO ₄
media					
Trasmission Range	1.1-6.5	0.38-1.8	0.38-5.5	0.35-5.5	0.2-5.3
(μm)					
Refractive index	3.42	1.556	1. 8 4 (o)	1.986	1.884 (o)
			1.711 (e)	p[mm]p	1.898 (e)
				2.033	
				p[gg]p	
Raman shift at 300K	521	1047.3	770	901	910.7
(cm ⁻¹)			822	768	
Spontaneous Raman	3.5	0.4	5.0	5.9	4.8
linewidth (cm ⁻¹)					
Raman gain	20	11	4.8	3.3	-
(cm/GW)	(1550nm)	(1064nm)	(1064nm)	(1064)	
Optical damage	~1000-4000	~400	~100	-	-
threshold (MW/cm ²)					
Thermal	148	1.17	-	2.6 [1 0 0]	16
conductivity				3.8 [0 1 0]	
(W/m-K)				3.4 [0 0 1]	

Silica glass



Continuum of vibration modes of the silica structure.

Maximum Raman gain is for the frequency offset of **13.2** THz.

A pump wave at **1064 nm** leads to the largest Raman gain at a Stokes wavelength of **1116 nm**.

Hydrogen gas, H_2 Raman shift =4155 cm⁻¹





1.064 µm laser pump

1st Stokes 1.91 μm 2nd Stokes 9.2 μm

A continuous-wave Raman silicon laser

A continuous-wave Raman silicon laser

2005

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Achieving optical gain and/or lasing in silicon has been one of the most challenging goals in silicon-based photonics^{1–3} because bulk silicon is an indirect bandgap semiconductor and therefore has a very low light emission efficiency. Recently, stimulated Raman scattering has been used to demonstrate light amplification and lasing in silicon^{4–9}. However, because of the nonlinear optical loss associated with two-photon absorption (TPA)-induced free

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carrier absorption (FCA)^{10–12}, until now lasing has been limited to pulsed operation^{8,9}. Here we demonstrate a continuous-wave silicon Raman laser. Specifically, we show that TPA-induced FCA in silicon can be significantly reduced by introducing a reversebiased p-i-n diode embedded in a silicon waveguide. The laser cavity is formed by coating the facets of the silicon waveguide with multilayer dielectric films. We have demonstrated stable single mode laser output with side-mode suppression of over 55 dB and linewidth of less than 80 MHz. The lasing threshold depends on the p-i-n reverse bias voltage and the laser wavelength can be tuned by adjusting the wavelength of the pump laser. The demonstration of a continuous-wave silicon laser represents a significant milestone for silicon-based optoelectronic devices. Silicon waveguide



Pump wavelength **1550 nm.** Raman/Stokes wavelength **1686 nm**

The effective core area $(1.6 \ \mu m)^2$ WG length: 4.8 cm

When a reverse bias voltage (~25V) is applied, the TPA-generated electron–hole pairs can be swept out of the silicon waveguide by the electric field between the p-and n-doped regions.

Raman gain was measured in a pump–probe experiment: a single-pass gain of 3 dB $(\sim 2^{x})$ at a pump power of 700 mW coupled into the waveguide.

A continuous-wave Raman silicon laser (continued)

NATURE | VOL 433 | 17 FEBRUARY 2005 | www.nature.com/nature © 2005 Nature 10.0 Pump power monitor Silicon 25 V bias Lensed waveguide ŦŦ Polarization 5 V bias fibre 8.0 controller Pump at De-multiplexer 25 V slope 1,550 nm Laser output (mW) 5 V slope 90/10 6.0 Tap coupler Laser I output at Dichroic High-4.0 1,686 nm coating reflection LP filter Optical coating spectrum 2.0 analyser 90/10 0.0 Tap coupler Laser output 200 400 600 800 0 power meter Coupled pump power (mW)

The Raman laser frequency shift is 15.6 THz (520 cm⁻¹) The lasing threshold was 180mW with a 25-V bias. Conversion efficiency 2% Figure 3 Silicon Raman laser output power as a function of the input pump power at a reverse bias of 25 and 5 V. The pump wavelength is 1,550 nm and the laser wavelength is
➡ 1,686 nm. The slope efficiency (single side output) is 4.3% for 25-V bias and 2% for 5-V bias. Error bars represent standard deviations.

monolithic integration of silicon-based optoelectronics

Cascaded Raman lasing



Cascaded Raman laser (converter) based on a tellurite fiber and Tm pump laser.

The Raman-active medium can be an optical fiber, a bulk crystal (e.g. silicon or diamond), a waveguide in a photonic integrated circuit, or a cell with a gas or liquid.

- Stokes wave can propagate in **any direction**, even counter-propagate
- In the telecom systems, **fiber Raman amplifiers** compete with erbiumdoped fiber amplifiers. Raman amplifiers can have co-propagating or counter-propagating pump wave (from continuous-wave diode laser).
- Raman amplifiers can be operated in different wavelength regions, provided that a suitable pump source is used.
- The gain spectrum can be tailored by using different pump wavelengths simultaneously.
- As opposed to SBS, pulsed (ns or ps) radiation can be used to reach SRS threshold
- Raman lasers can be used as frequency down-converters





Signal power vs. distance with EDFA only

Signal power vs.distance with EDFA and Raman amplifier

Raman frequency self-shift

Soliton self-frequency shift. Due to Raman gain, the **blue portion of the soliton spectrum pumps the red portion of the spectrum**, causing a continuous redshift in the soliton spectrum.



Coherent Anti-Stokes Raman Scattering (CARS)

Coherent Anti-Stokes Raman Scattering (CARS)

Can be regarded as a resonant 4-wave mixing process

4-wave process

 $\omega_{AS} = \omega_L - \omega_S + \omega_L = 2\omega_L - \omega_S$

CARS needs a strong Stokes wave

Phase matching is critical

$$k_{AS} = 2k_L - k_S \tag{23.5}$$

(23.4)



Collinear phase matching is impossible for normal dispersion, but vector phase matching - is possible; hence the outputs are in the form of a cone.





FIGURE 10.3.5 Phase-matching relations for Stokes and anti-Stokes coupling in stimulated Raman scattering.

CARS microscopy









20 µm



Comparison of label-free CAR... researchgate.net





