PhD Seminars – Season 7 Episode 2

An insight into the wonderful world of polarization ultrafast spectroscopy

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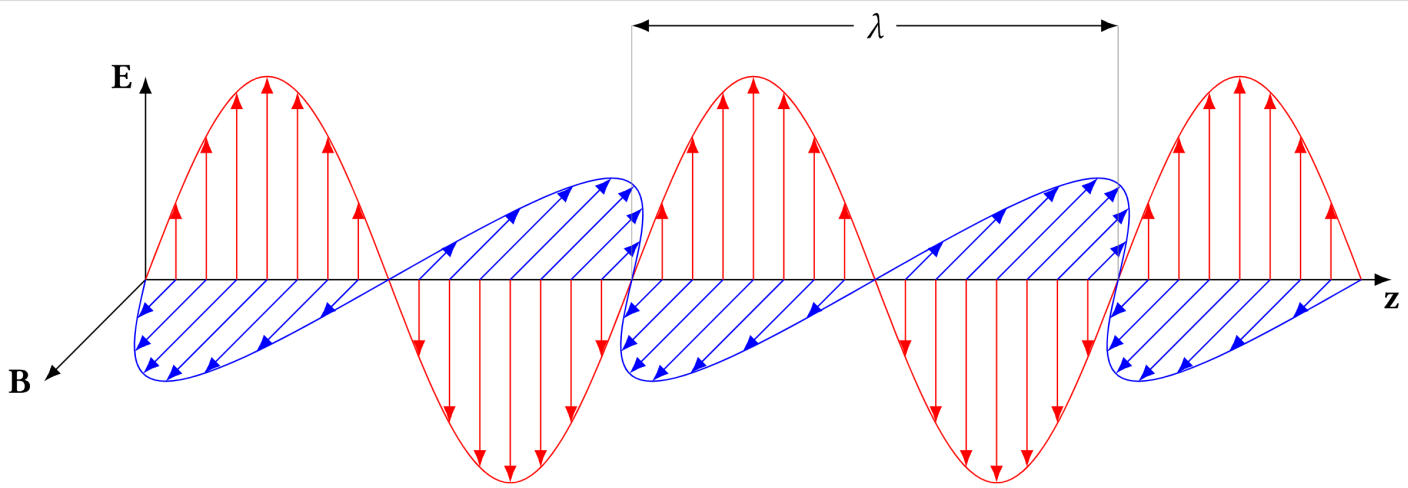
Summary

- What is light polarization?
- Kerr effect
- Raman effect
- (Raman + Kerr) Effect = Raman induced Kerr effect

Polarization

A word enclosing different meanings in physics and in other fields



Electromagnetic wave:

direction of the oscillations of the electric field during the wave propagation

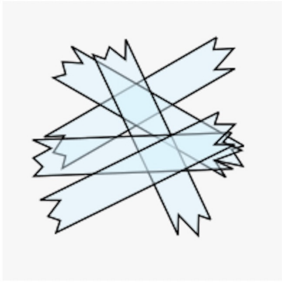
Abstract art?

No, linearly polarized light passing through a clear surface cover with clear tape

Polarization

How to obtain an abstract painting with polarized light

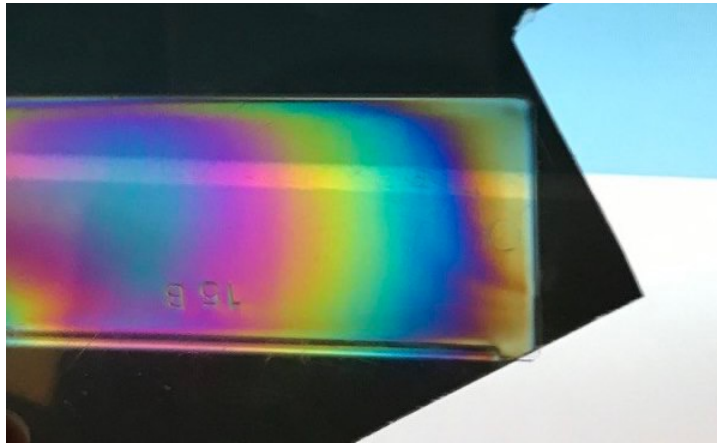
1. Cover a clear surface with tape



2. Take a source of polarized light (an LCD monitor)

3. Take polarizer (a pair of sunglasses)

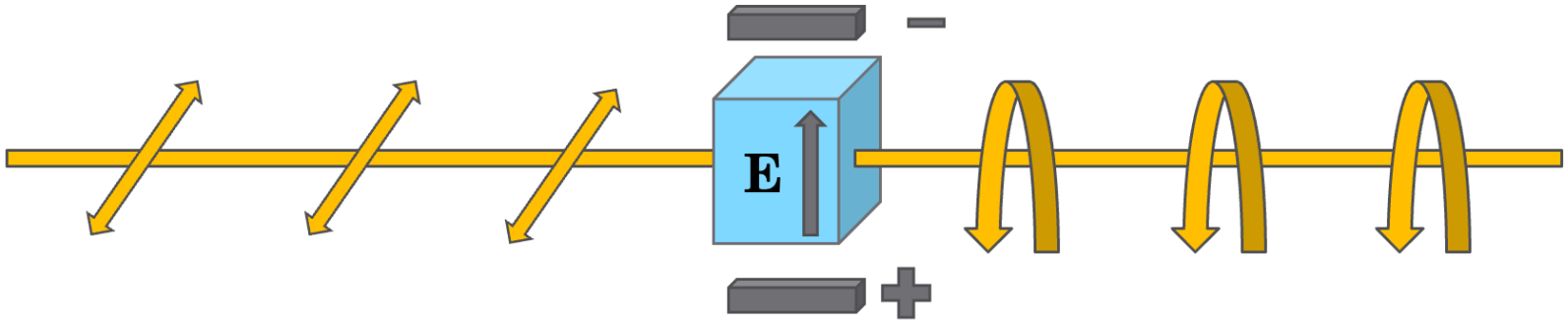
4. Properly combining the three object you are ready for the GNAM



Kerr Effect (elettro-optic)

Kerr discover in 1875

Electric field induces anisotropy in the refractive index



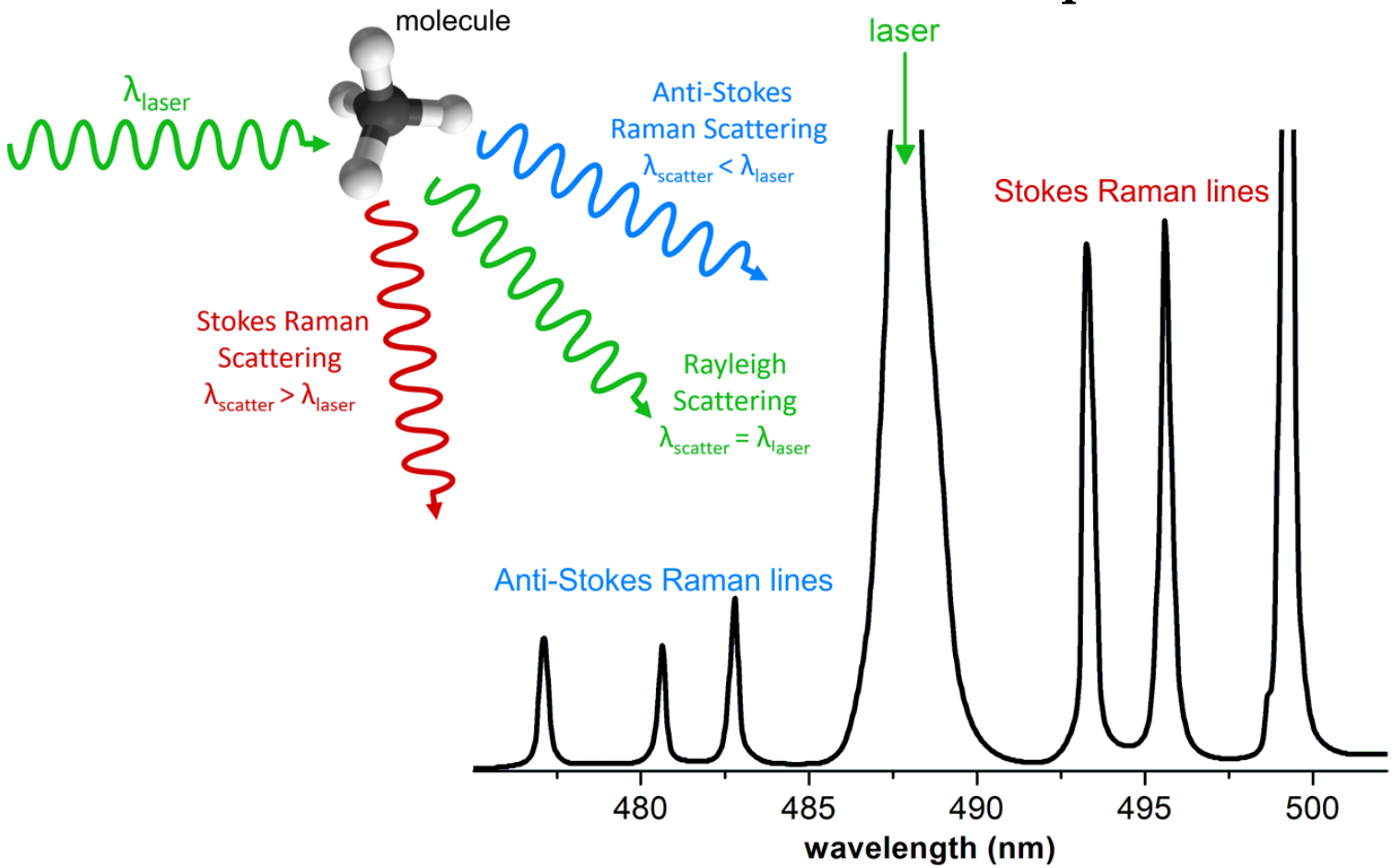
$$n_{\parallel} - n_{\perp} \propto |E_0|^2$$

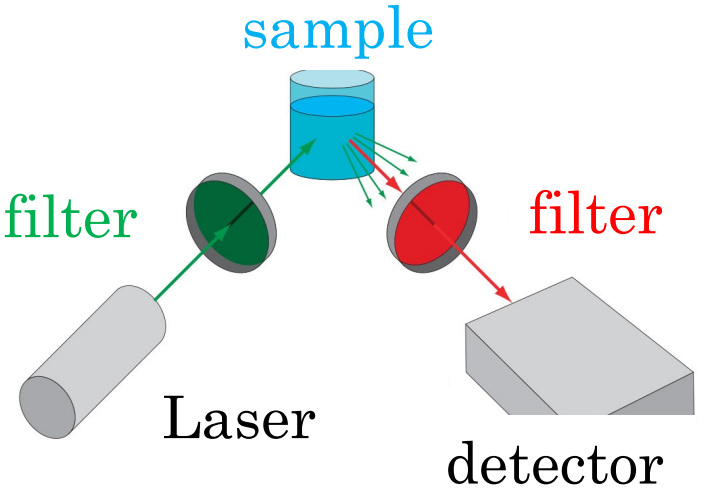
Components of light beam along the two axis experience different refractive indexes acquiring a relative phase

Non linear effect in the electric field strength

Raman Effect

Light – matter inelastic scattering process resulting in a **frequency shift** of the incident light of a **vibrational quantum**

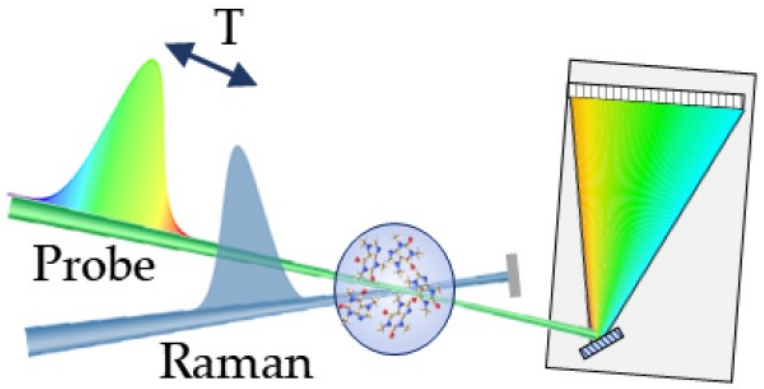




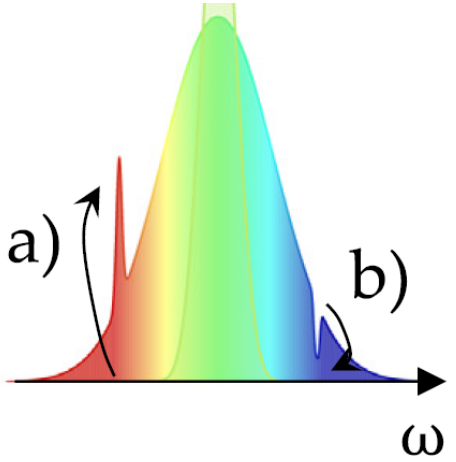
Stimulated Raman Spectroscopy

Two pulsed beams non linear interaction generates a

vibrational coherence



- Signal as modification of the probe spectral shape
- **Photoinduced dynamics** on ultrashort time scales



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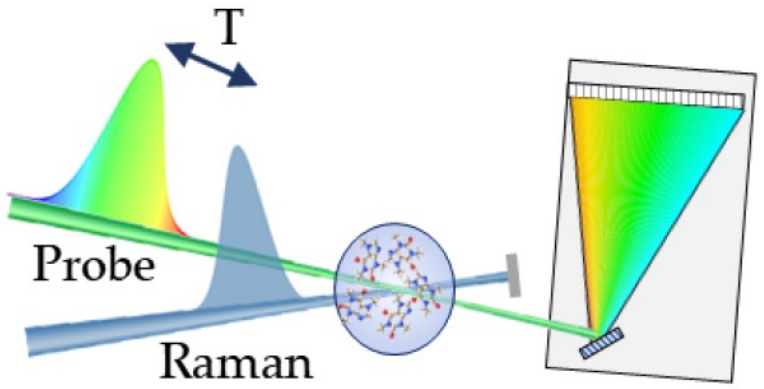

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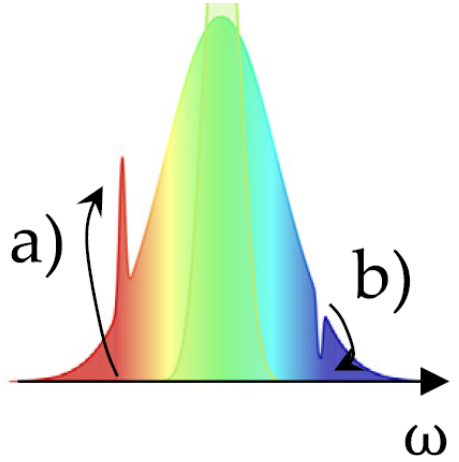
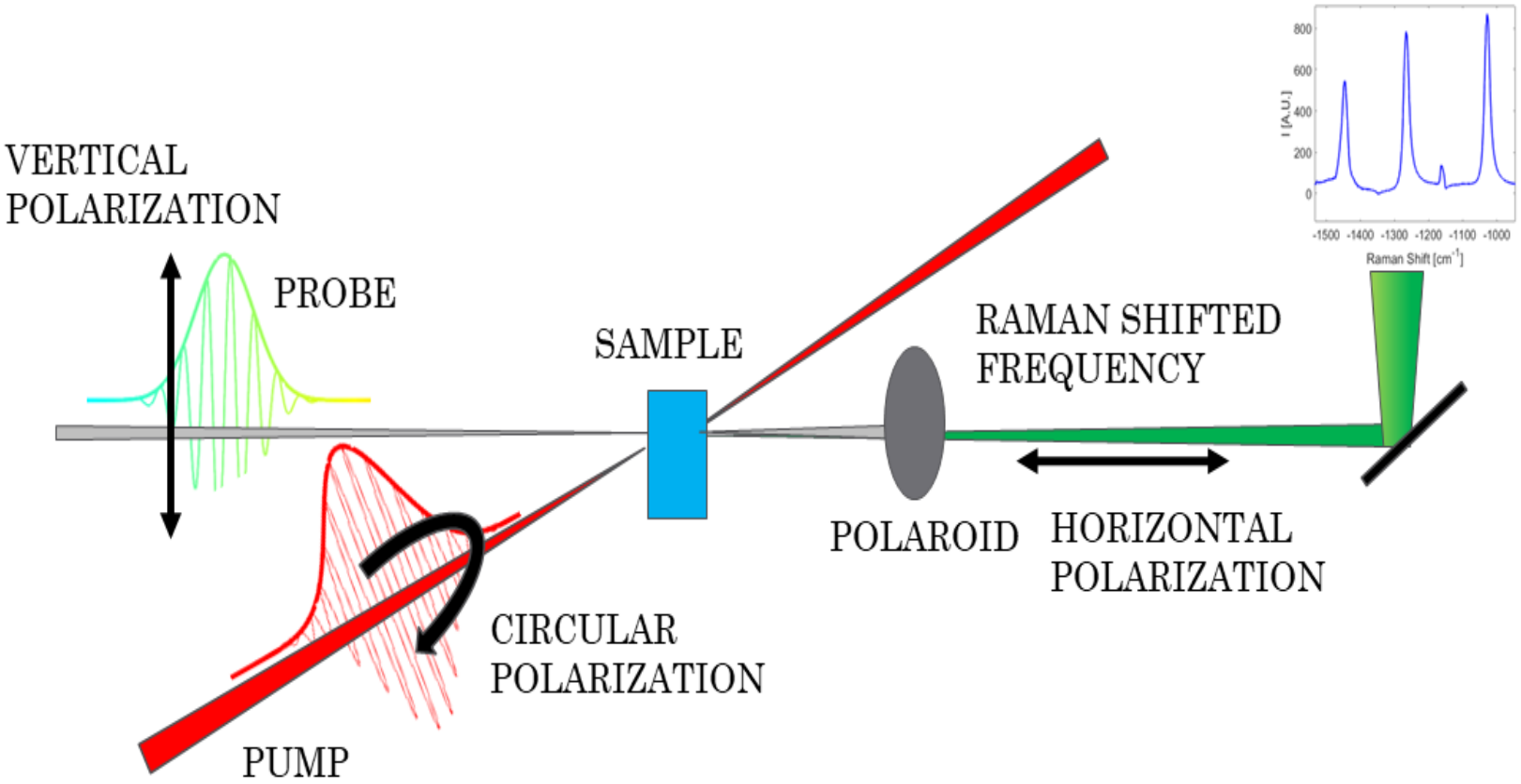


$$\nabla^2 \boldsymbol{E} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{E} = -\mu_0 \frac{\partial^2}{\partial t^2}\boldsymbol{P}$$

$$\boldsymbol{P} = \boldsymbol{\chi E} + \boldsymbol{\chi^{(2)} E^2} + \boldsymbol{\chi^{(3)} E^3}$$

$$= \boldsymbol{P^{(1)}} + \boldsymbol{P^{(2)}} + \boldsymbol{P^{(3)}}$$

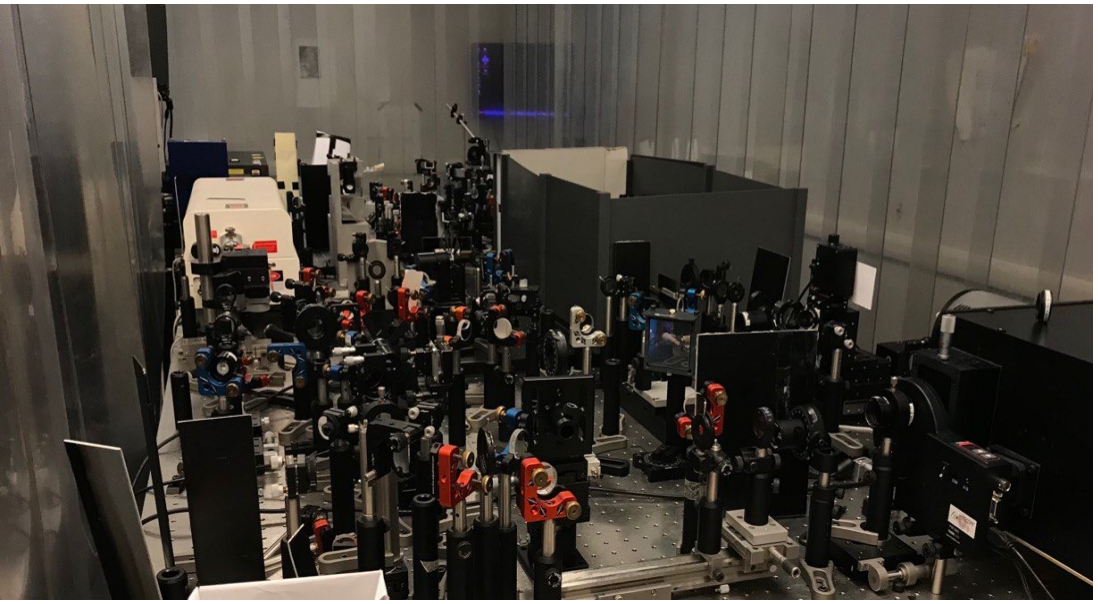
Raman Induced Kerr Effect

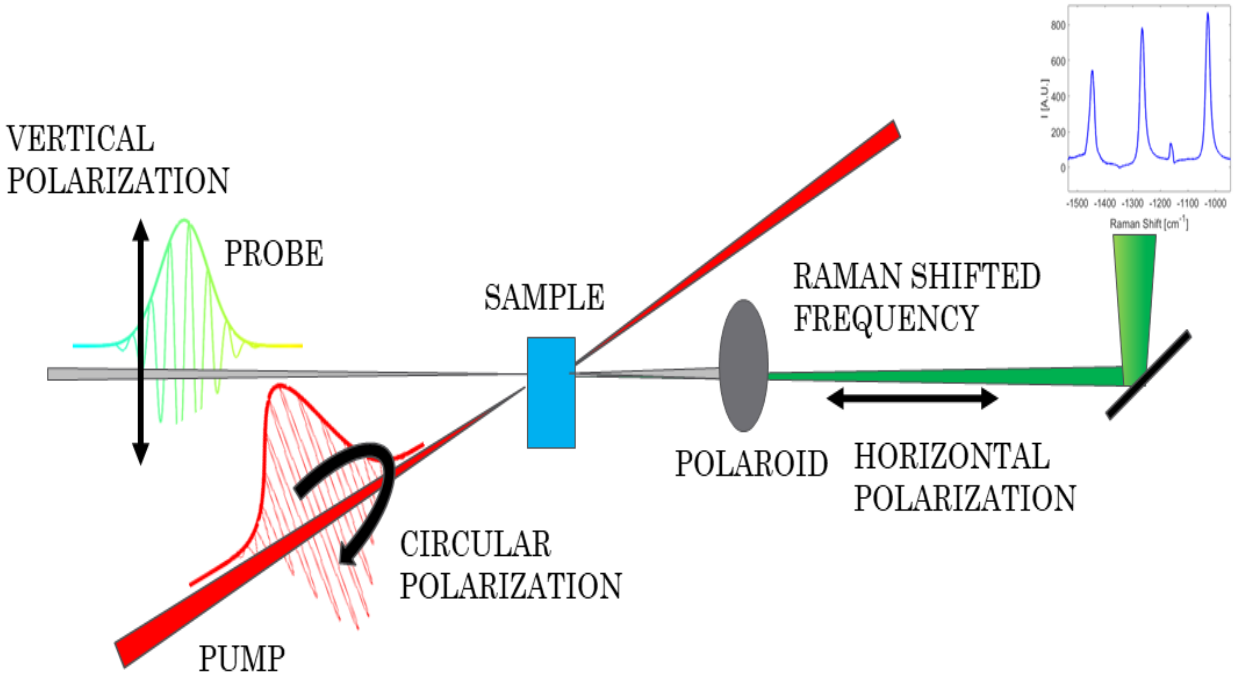


Birifrangence induced at Raman shifted frequencies

Raman Induced Kerr Effect

Birifrangence induced at Raman shifted frequencies





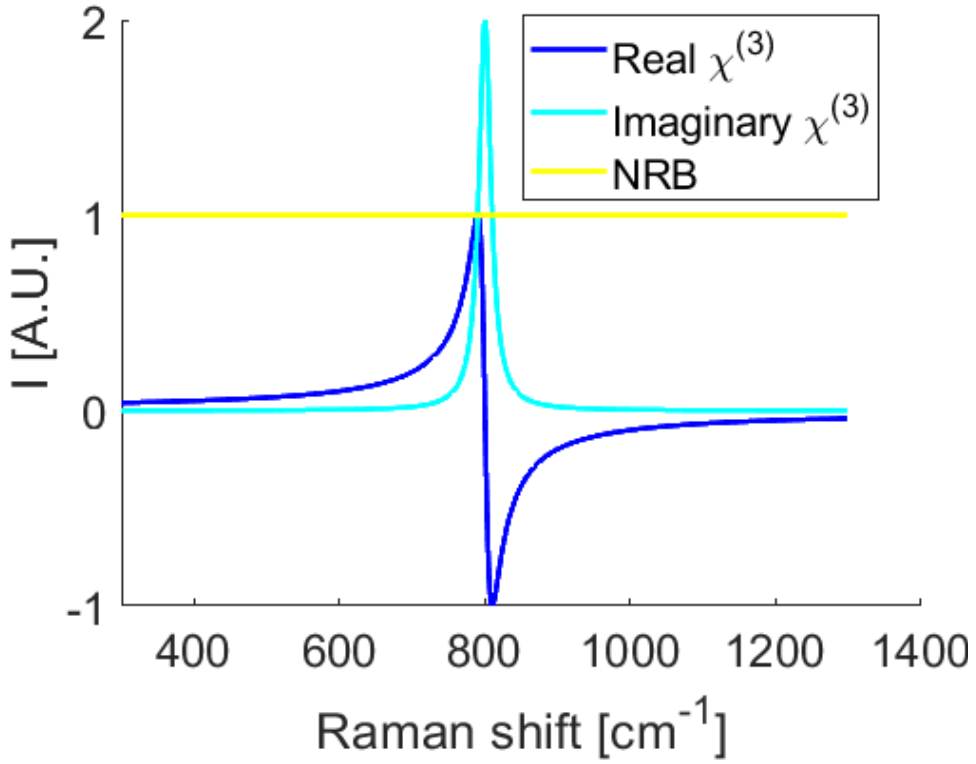
Stimulated Raman scattering based on the beams

polarization with detection **orthogonal** to the **probe**

$$P_x^{(3)}(\omega) \propto \chi_{xyxy}^{(3)}(\omega;\omega_R,-\omega_R,\omega_P)E_y^R(E_x^R)^*E_P + \chi_{xxyy}^{(3)}(\omega;\omega_R,-\omega_R,\omega_P)\,E_x^R(E_y^R)^*E_P$$

RIKE spectra in the Non Resonant case

$$P_x^{(3)}(\omega) \propto \chi^{(3)}_{xyxy} E_y^R (E_x^R)^* E_y^P + \chi^{(3)}_{xxyy}\, E_x^R (E_y^R)^* E_y^P$$



- Raman Non linear susceptibility

$$\chi^{(3)} = \frac{N\,\sigma}{\omega_v - \omega + i\,\Gamma}$$

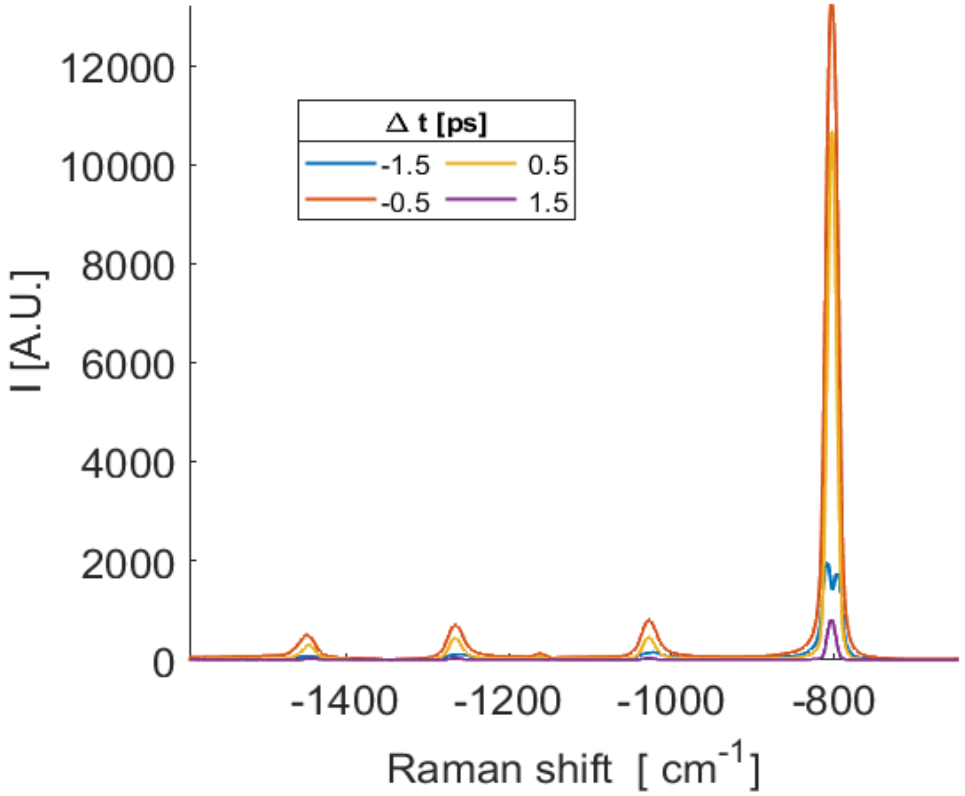
- Frequency independent Non Resonant Background

$$^{NRB}\chi^{(3)}_{xyxy} = {}^{NRB}\chi^{(3)}_{xxyy}$$

A circular polarized pump suppresses the non resonant background and selects the imaginary part of non linear Raman susceptibility

RIKE spectra in the Non Resonant case

Circular polarized pump



$$P_x^{(3)}(\omega) \propto i\,(\chi^{(3)}_{xyxy} - \chi^{(3)}_{xxyy})\, I_R E_P$$

- Raman **spectrum free** from any background, linear and non linear
- Elevated signal to noise ratio
- Access to off-diagonal element of non linear susceptibility

Summary

- The polarization of light is a fundamental degree of fredoom of light: exploited in many applications, it originates many wonderful phenomena
- **Raman Induced Kerr Effect** allows to record **background free high signal to noise ratio** Raman spectra