Quantum model for Impulsive Stimulated Raman Scattering (ISRS)

Stefano Marcantoni

University of Trieste

Trieste Junior Quantum Days
May 18, 2018

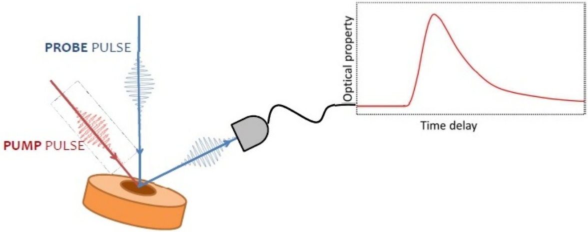
Outline

INCEPT

INhomogenieties and fluctuations in quantum CohErent Phases by ultrafast optical Tomography

- Experiments: Prof. Daniele Fausti (P.I.),
  Theory: Prof. Fabio Benatti
- Ultrashort dynamics in complex materials (sub-picosecond time scales)
- Cross-fertilization between quantum optics (quantum state tomography) and condensed matter physics (pump-probe experiments)

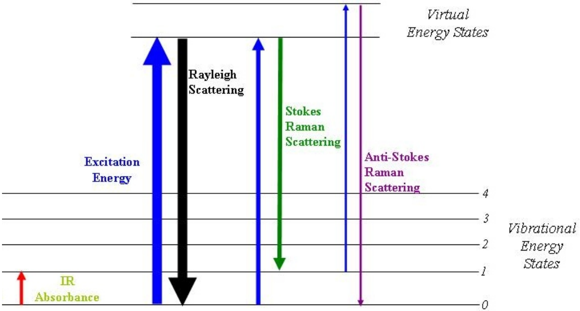
Pump-probe experiments



- Pump pulse excites the material
- Probe pulse (less intense) test the evolution after a delay time t

Raman scattering

- Raman scattering is a kind of inelastic scattering for light
- One photon loses energy exciting one phonon in the material (Stokes process)... or
  One photon increases its energy destroying one phonon (anti-Stokes process)



Probe-target interaction

- Initial state (probe + target): $|\alpha\rangle\langle\alpha| \otimes \varrho_t$
- Refraction at the boundary

$$H_{ref} = \sum_{j,\mu,\mu'} \left(\eta^{(0)}_{\mu\mu'} + \langle b + b^\dagger\rangle_t \, \eta^{(1)}_{\mu\mu'}\right)\left(a^\dagger_{\mu j}\, r_{\mu' j} + a_{\mu j}\, r^\dagger_{\mu' j}\right)$$

- Raman scattering

$$H_{Ram} := \sum_{\mu,\mu'} \chi_{\mu,\mu'} \left[\left(\sum_j a^\dagger_{\mu j} a_{\mu' j+\frac{\Omega}{\delta}}\right) b^\dagger + \left(\sum_j a_{\mu j} a^\dagger_{\mu' j+\frac{\Omega}{\delta}}\right) b\right],$$

$a_{\mu j}, r_{\mu j}, b$ are bosonic operators

Dynamics and observables

- Evolution operator

$$U(\tau) = U_{bulk}(\tau)\, U_{ref}$$

$$U_{ref} = \exp(-i\, H_{ref})\ , \quad U_{bulk}(\tau) = \exp(-i\tau\, H_{Raman}) \qquad (\hbar = 1)\ ,$$

- Average of an observable X_{phot}

$$\langle X_{phot}(\tau)\rangle_t = \mathrm{Tr}[U(\tau)\, |\alpha\rangle\langle\alpha| \otimes \varrho_t\, U^\dagger(\tau)\, X_{phot}]$$

- Coherent state of the probe

$$|\alpha\rangle = \exp\left(\sum_j \alpha_{xj} a^\dagger_{xj} - \alpha^*_{xj} a_{xj}\right)|0\rangle\ , \quad a_{xj}|\alpha\rangle = \alpha_{xj}\ , \quad a_{yj}|\alpha\rangle = 0\ ,$$

$$\alpha_{xj} = \exp\left(-\frac{(j\delta)^2}{2\sigma^2}\right) \mathrm{e}^{i\varphi}$$

Pump-target interaction

- Same Hamiltonian for the light-matter interaction but different approximation
- Mean field for photons $a_{\lambda j} \to \alpha^P_{\lambda j}$
- Explicit dependence on the polarization angle (with respect to x)

$$\alpha^P_{xj} = \alpha^P_{0j} \cos(\theta_P), \quad \alpha^P_{yj} = \alpha^P_{0j} \sin(\theta_P)$$

- Phononic operator shifted $\langle b \rangle_t = \mathrm{Tr}(\varrho_t b) = \mathrm{Tr}(\varrho b_t)$

$$\begin{aligned} b \to b_t =& U^\dagger_{ref}\, U^\dagger_{bulk}(\tau)\, U^\dagger_{free}(t)\, b\, U_{free}(t)\, U_{bulk}(\tau)\, U_{ref} \\ \simeq& \, \mathrm{e}^{-i\Omega t} \left(b - i\tau \sum_{j,\lambda\lambda'} \chi_{\lambda\lambda'}\, \alpha^{P*}_{\lambda j}\, \alpha^P_{\lambda' j+\frac{\Omega}{\delta}} \right) . \end{aligned}$$

Assumptions on the interaction (good for Quartz)

- Zeroth order refraction matrix

$$\eta^{(0)} = \begin{pmatrix} \eta_{xx}^{(0)} & \eta_{xy}^{(0)} \\ \eta_{xy}^{(0)} & \eta_{xx}^{(0)} \end{pmatrix}$$

- First order refraction matrix depending on the phonon involved (same for χ)

$$A : \eta^{(1)} = \begin{pmatrix} \eta_{xx}^{(1)} & 0 \\ 0 & \eta_{xx}^{(1)} \end{pmatrix}, \quad E_L : \eta^{(1)} = \begin{pmatrix} \eta_{xx}^{(1)} & 0 \\ 0 & -\eta_{xx}^{(1)} \end{pmatrix},$$

$$E_T : \eta^{(1)} = \begin{pmatrix} 0 & \eta_{xy}^{(1)} \\ \eta_{xy}^{(1)} & 0 \end{pmatrix}$$

Geometry

- Phonons selected by the angle between the polarization of the pump and the x axis (θ_P)

$$\begin{aligned} A :\langle b\rangle_t &= C_A \mathrm{e}^{-i\Omega_A t - i\pi/2} \\ E_L :\langle b\rangle_t &= C_E \cos(2\theta_P)\, \mathrm{e}^{-i\Omega_E t - i\pi/2}\ , \\ E_T :\langle b\rangle_t &= C_E \sin(2\theta_P)\, \mathrm{e}^{-i\Omega_E t - i\pi/2}\ , \end{aligned}$$

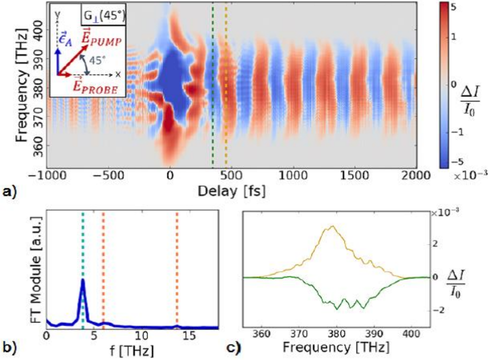
Remember:

$$\begin{aligned} b \rightarrow & U^\dagger_{ref}\, U^\dagger_{bulk}(\tau)\, U^\dagger_{free}(t)\, b\, U_{free}(t)\, U_{bulk}(\tau)\, U_{ref} \\ & \simeq \mathrm{e}^{-i\Omega t} \left(b - i\tau \sum_{j,\lambda\lambda'} \chi_{\lambda\lambda'}\, \alpha^{P*}_{\lambda j}\, \alpha^{P}_{\lambda' j + \frac{\Omega}{\delta}} \right) . \end{aligned}$$

Mode occupation numbers (y polarization)

$$\langle N_{yk}(\tau)\rangle_t \simeq \langle b + b^\dagger\rangle_t |\alpha_{xk}|^2 \, F^y_{ref} - i\,\langle b^\dagger - b\rangle_t |\alpha_{xk}| \left(|\alpha_{xk+\frac{\Omega}{\delta}}| - |\alpha_{xk-\frac{\Omega}{\delta}}|\right) F^y_{Ram}(\tau)$$

$$\begin{aligned} A: &\quad F^y_{ref} = 0, && F^y_{Ram}(\tau) = 0, \\ E_L: &\quad F^y_{ref} = 0, && F^y_{Ram}(\tau) = 0, \\ E_T: &\quad F^y_{ref} = 2\eta^{(1)}_{xy}\eta^{(0)}_{xy}\sin^2(\eta^{(0)}_{xx}), && F^y_{Ram}(\tau) = 0. \end{aligned}$$

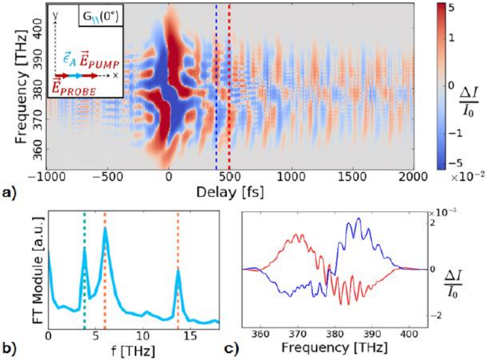


- Pump at 45°: A and E_T phonons excited
- Orthogonal polarization (leading term): E_T Refractive

Mode occupation numbers (x polarization)

$$\langle N_{xk}(\tau)\rangle_t \simeq \cos^2(\eta_{xx}^{(0)})|\alpha_{xk}|^2 + \langle b+b^\dagger\rangle_t|\alpha_{xk}|^2\, F_{ref}^x \\ - i\,\langle b^\dagger - b\rangle_t|\alpha_{xk}|\left(|\alpha_{xk+\frac{\Omega}{\delta}}| - |\alpha_{xk-\frac{\Omega}{\delta}}|\right)\, F_{Ram}^x(\tau)$$

$$\begin{aligned} A:&\quad F_{ref}^x = -\eta_{xx}^{(1)}\sin(2\eta_{xx}^{(0)}), && F_{Ram}^x(\tau) = \chi_{xx}\tau\cos^2(\eta_{xx}^{(0)}),\\ E_L:&\quad F_{ref}^x = -\eta_{xx}^{(1)}\sin(2\eta_{xx}^{(0)}), && F_{Ram}^x(\tau) = \chi_{xx}\tau\cos^2(\eta_{xx}^{(0)}),\\ E_T:&\quad F_{ref}^x = -2\eta_{xy}^{(1)}\eta_{xy}^{(0)}\cos^2(\eta_{xx}^{(0)}), && F_{Ram}^x(\tau) = 0. \end{aligned}$$

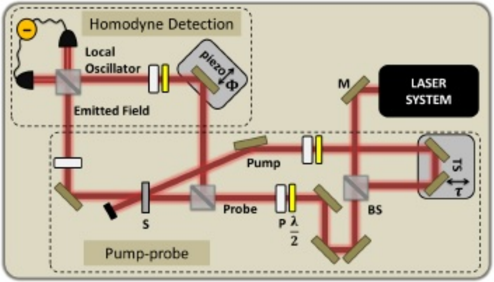


- Pump at 0°: A and E_L phonons excited
- Parallel polarization: Raman and Refractive effects are both visible

Results Experiment 1 (summary)

- Phase mismatch between Raman and refractive modulation
- Selection of different phonons according to the polarization of the pump
- Different behaviour of Raman and refractive modulation depending on the phonon involved and on the polarization selected by the analyzer
- Good agreement between theory and experiment

Quadrature: Homodyne detection + Time-resolved spectroscopy



- We combine two different experimental techniques to probe the nonequilibrium response of the material

Average quadrature

- Measured quantity: Current difference I

$$I = \sum_j \left(c_{xj}^\dagger c_{xj} \; - \; d_{xj}^\dagger d_{xj} \right) , \qquad c_j = \frac{a_{xj} + a_{xj}^{LO}}{\sqrt{2}} \, , \; d_j = \frac{a_{xj} - a_{xj}^{LO}}{\sqrt{2}} \; .$$

- Quadrature:

$$X_s = \frac{1}{\sqrt{2}} \sum_j \left(a_{xj} \, z_j^* \, \mathrm{e}^{-i\Phi_j(s)} \; + \; a_{xj}^\dagger \, z_j \, \mathrm{e}^{i\Phi_j(s)} \right) \propto I$$

- Theoretical prediction:

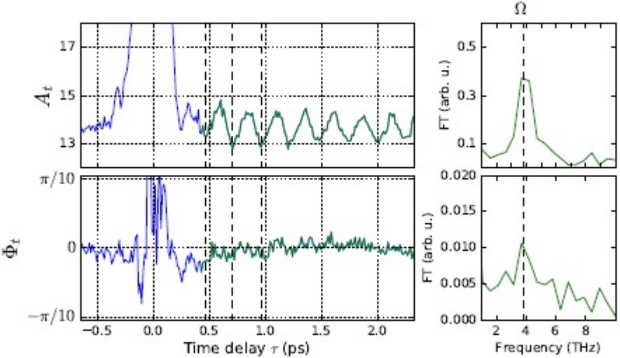
$$\langle X_s(\tau) \rangle = \mathcal{A}_t \cos(\omega_0 s + \Phi_t)$$

where

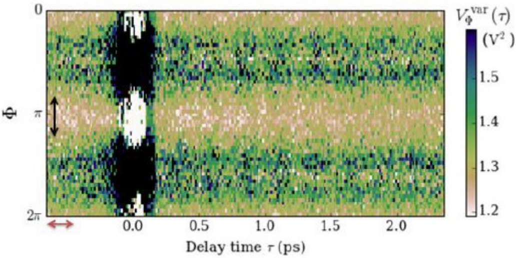
$$\mathcal{A}_t \simeq \mathcal{A} \left(1 + \overline{\eta} \sin(\Omega t)\right) , \quad \Phi_t \simeq 2\overline{\chi} \sin(\Omega t).$$

Average quadrature

$$\mathcal{A}_t \simeq \mathcal{A}\left(1 + \overline{\eta}\sin(\Omega t)\right), \quad \Phi_t \simeq 2\overline{\chi}\sin(\Omega t).$$



Variance of the quadrature: work in progress



- For a coherent initial state: variance is time-independent up to second order in the coupling
- Higher order effects or (more likely) signature of a statistical mixture

Conclusions and Outlook

Results:

- Fully quantum model for Impulsive Stimulated Raman Scattering (ISRS)
- Outcomes of two different experiments correctly reproduced

Future work:

- Complete tomography of the state of light (variance of the quadrature)
- Role of quantum correlations
- More interesting (complex) dynamics in the sample (*e.g.* interaction between the vibrational and electronic degrees of freedom)

Thank you for your attention!

Social dinner: Pizzeria "Al Barattolo" at 20.00.