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Group Seminar

June 2, 2006

Heterodyne CARS imaging: First results

Vishnu Vardhan Krishnamachari

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CARS is not Raman!

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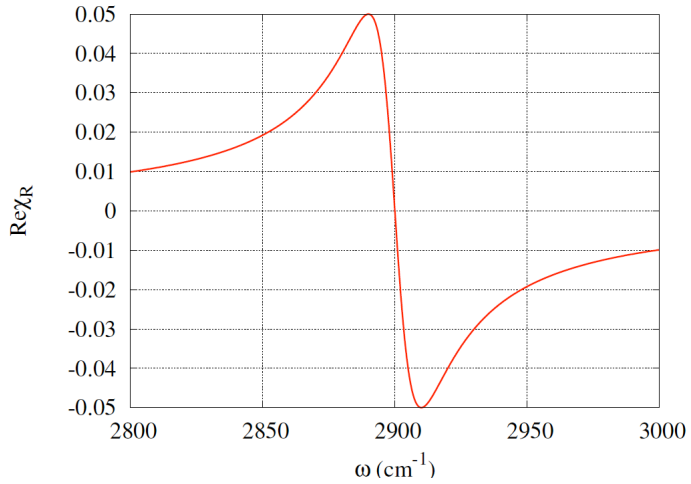
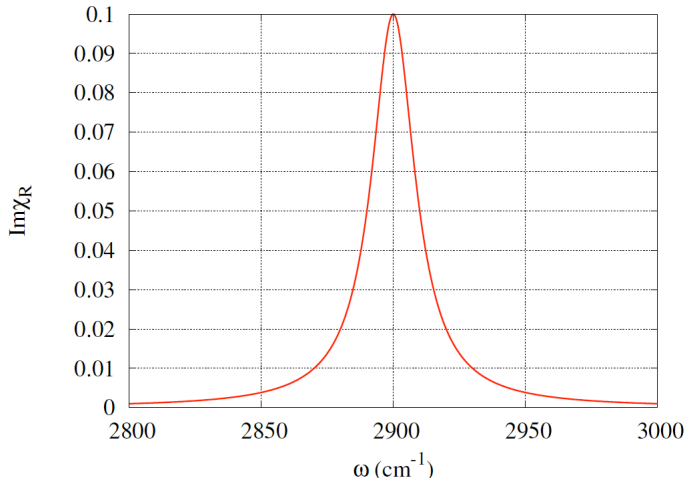
Frequency dependence of third-order non-linear susceptibility around a Raman transition

$$\chi_R^{(3)}(\omega) \propto -\frac{1}{\omega - \omega_0 + i\Gamma_0} \qquad \omega = \omega_p - \omega_S$$

$$= -\frac{\omega - \omega_0}{(\omega - \omega_0)^2 + \Gamma_0^2} + i\frac{\Gamma_0}{(\omega - \omega_0)^2 + \Gamma_0^2}$$

$$= \mathrm{Re}\left\{\chi_R^{(3)}\right\} + i\,\mathrm{Im}\left\{\chi_R^{(3)}\right\}$$





CARS is not Raman!

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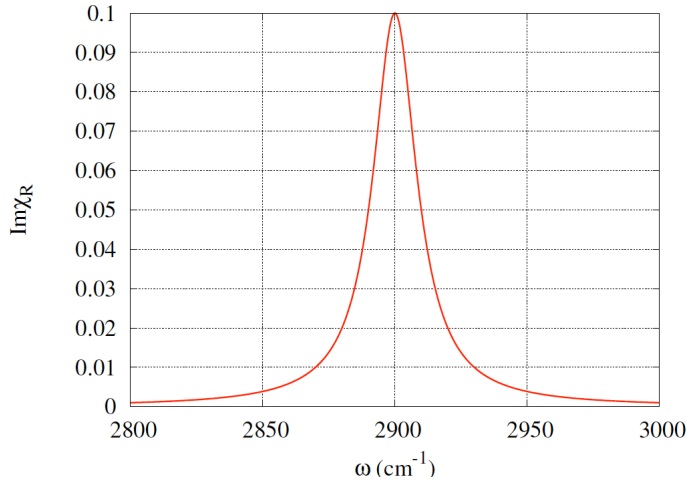
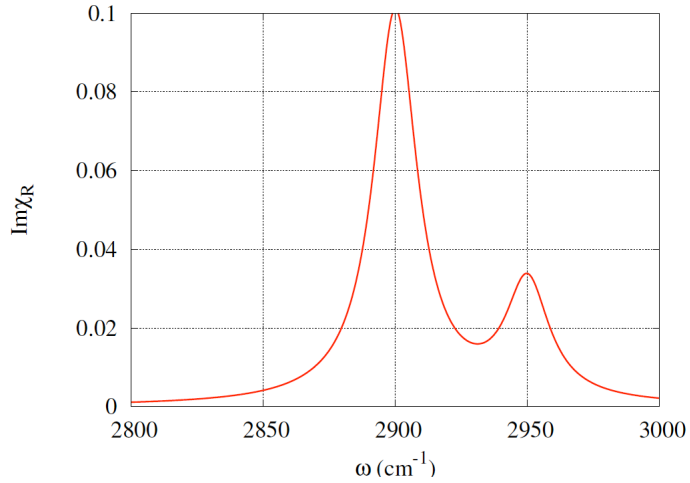
Spontaneous Raman emission intensity

$$I_{Raman} \propto \mathcal{N} \cdot \mathrm{Im}\left\{\chi_R^{(3)}\right\}$$

$$\propto \mathcal{N} \cdot \frac{\Gamma_0}{(\omega - \omega_0)^2 + \Gamma_0^2}$$





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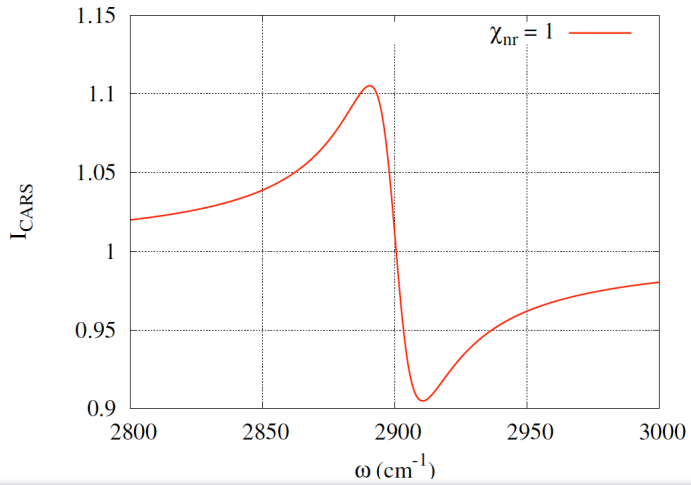
CARS intensity : Single peak

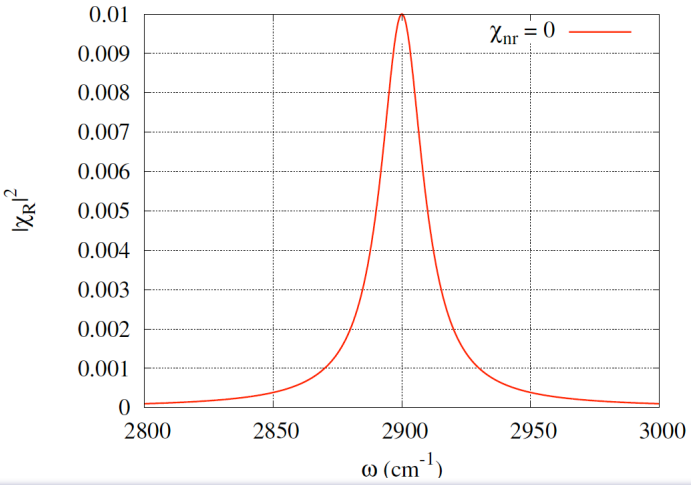
$$A_{CARS}(\omega) \propto \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \chi_R^{(3)}$$

$$I_{CARS}(\omega) \propto \left| \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \chi_R^{(3)} \right|^2$$

$$\propto \left| \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \mathrm{Re}\left\{\chi_R^{(3)}\right\} + i\mathcal{N} \cdot \mathrm{Im}\left\{\chi_R^{(3)}\right\} \right|^2$$

$$\propto \mathcal{N}'^2 \cdot \left(\chi_{nr}^{(3)}\right)^2 + \mathcal{N}^2 \cdot \left|\chi_R^{(3)}(\omega)\right|^2 + 2\mathcal{N}' \cdot \mathcal{N} \cdot \chi_{nr}^{(3)} \cdot \mathrm{Re}\left\{\chi_R^{(3)}(\omega)\right\}$$





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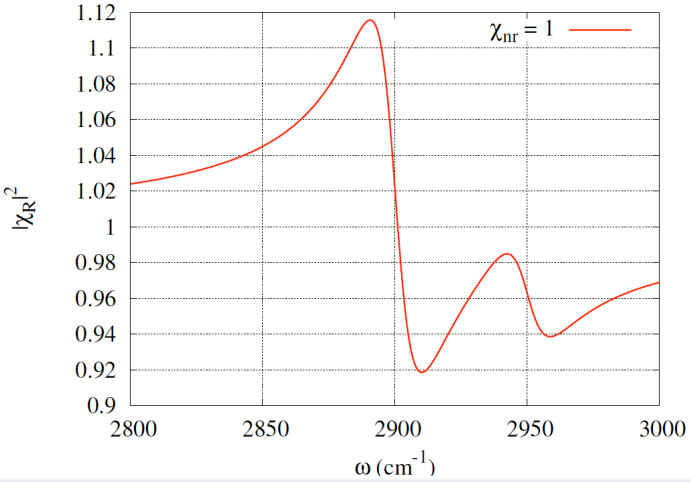
CARS intensity : Double peak

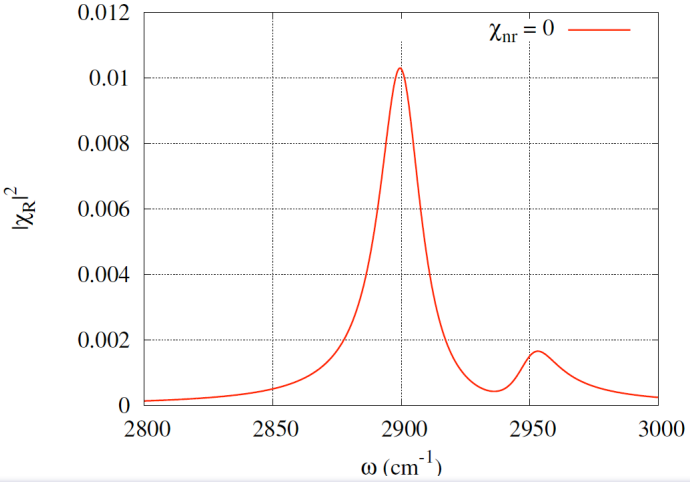
$$A_{CARS}(\omega) \propto \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \chi_R^{(3)}$$

$$I_{CARS}(\omega) \propto \left| \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \chi_R^{(3)} \right|^2$$

$$\propto \left| \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \mathrm{Re}\left\{\chi_R^{(3)}\right\} + i\mathcal{N} \cdot \mathrm{Im}\left\{\chi_R^{(3)}\right\} \right|^2$$

$$\propto \mathcal{N}'^2 \cdot \left(\chi_{nr}^{(3)}\right)^2 + \mathcal{N}^2 \cdot \left|\chi_R^{(3)}(\omega)\right|^2 + 2\mathcal{N}' \cdot \mathcal{N} \cdot \chi_{nr}^{(3)} \cdot \mathrm{Re}\left\{\chi_R^{(3)}(\omega)\right\}$$





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CARS intensity

$$A_{CARS}(\omega) \propto \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \chi_R^{(3)}$$

$$I_{CARS}(\omega) \propto \left| \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \chi_R^{(3)} \right|^2$$

$$\propto \left| \mathcal{N}' \cdot \chi_{nr}^{(3)} + \mathcal{N} \cdot \mathrm{Re}\left\{\chi_R^{(3)}\right\} + i\mathcal{N} \cdot \mathrm{Im}\left\{\chi_R^{(3)}\right\} \right|^2$$

$$\propto \mathcal{N}'^2 \cdot \left(\chi_{nr}^{(3)}\right)^2 + \mathcal{N}^2 \cdot \left|\chi_R^{(3)}(\omega)\right|^2 + 2\mathcal{N}' \cdot \mathcal{N} \cdot \chi_{nr}^{(3)} \cdot \mathrm{Re}\left\{\chi_R^{(3)}(\omega)\right\}$$

- Non-resonant and resonant signals interfere
- Difficult to analyze the spectrum
- Huge background => Poor SNR & sensitivity
- CARS intensity is not proportional to the concentration

Heterodyne CARS

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Heterodyne detection:
Detection of the desired signal by mixing it with a "known" or "well-characterized" signal.

$$
\begin{aligned}
A_{\mathrm{Het}} &\propto \mathcal{N}' \cdot \chi^{(3)}_{nr} + \mathcal{N} \cdot \chi^{(3)}_{R} + \chi^{(3)}_{lo} \\
&\propto \left(\mathrm{Re}\left\{\chi^{(3)}_{lo}\right\} + \mathcal{N}' \cdot \chi^{(3)}_{nr} + \mathcal{N} \cdot \mathrm{Re}\left\{\chi^{(3)}_{R}\right\}\right) \\
&\quad + i\left(\mathrm{Im}\left\{\chi^{(3)}_{lo}\right\} + \mathcal{N} \cdot \mathrm{Im}\left\{\chi^{(3)}_{R}\right\}\right)
\end{aligned}
$$

$$
\begin{aligned}
I_{\mathrm{Het}} &\propto 2\mathrm{Re}\left\{\chi^{(3)}_{lo}\right\}\left(\mathcal{N}' \cdot \chi^{(3)}_{nr} + \mathcal{N} \cdot \mathrm{Re}\left\{\chi^{(3)}_{R}\right\}\right) \\
&\quad + 2\mathcal{N} \cdot \mathrm{Im}\left\{\chi^{(3)}_{lo}\right\} \cdot \mathrm{Im}\left\{\chi^{(3)}_{R}\right\} + \cdots
\end{aligned}
$$

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Heterodyne coherent anti-Stokes Raman scattering (CARS) imaging

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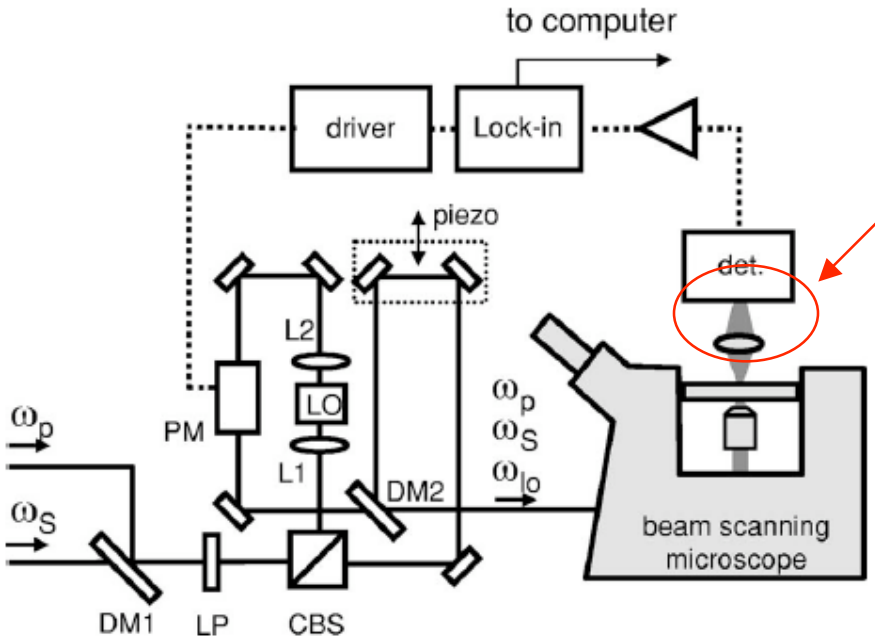
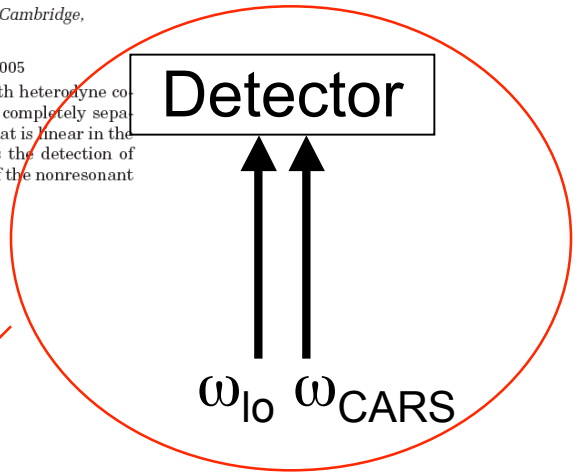
We have achieved rapid nonlinear vibrational imaging free of nonresonant background with heterodyne coherent anti-Stokes Raman scattering (CARS) interferometric microscopy. This technique completely separates the real and imaginary responses of nonlinear susceptibility $\chi^{(3)}$ and yields a signal that is linear in the concentration of vibrational modes. We show that heterodyne CARS microscopy permits the detection of weak vibrational resonances that are otherwise overshadowed by the strong interference of the nonresonant background. © 2006 Optical Society of America

OCIS codes: 180.6900, 190.5650





- $\Delta\phi = \pi/2$
  $\Rightarrow I_{out} \propto \mathrm{Im}\{\chi^{(3)}\}$
  $\propto$ Conc
- $\Delta\phi = 0$
  $\Rightarrow I_{out} \propto \mathrm{Re}\{\chi^{(3)}\}$

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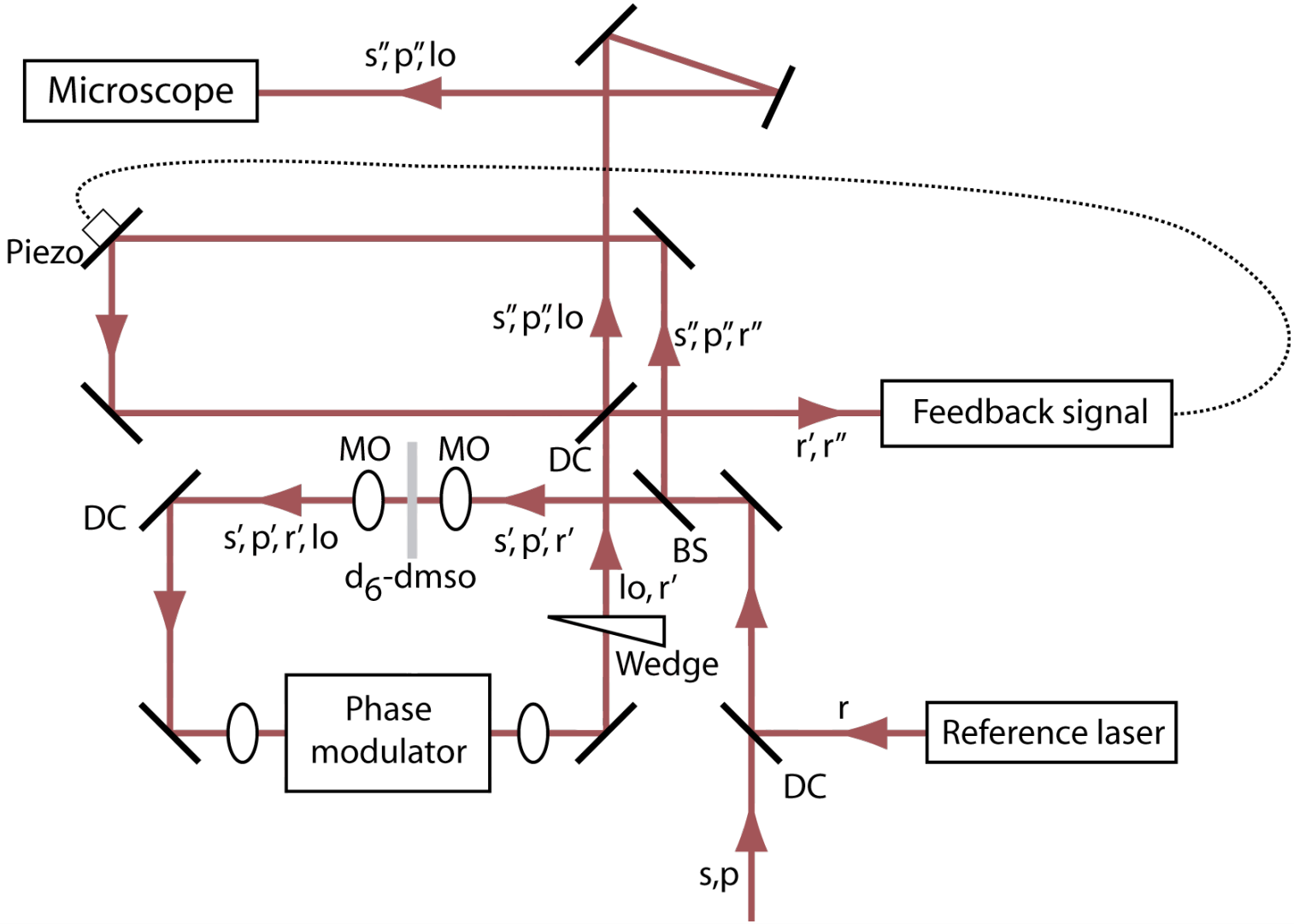

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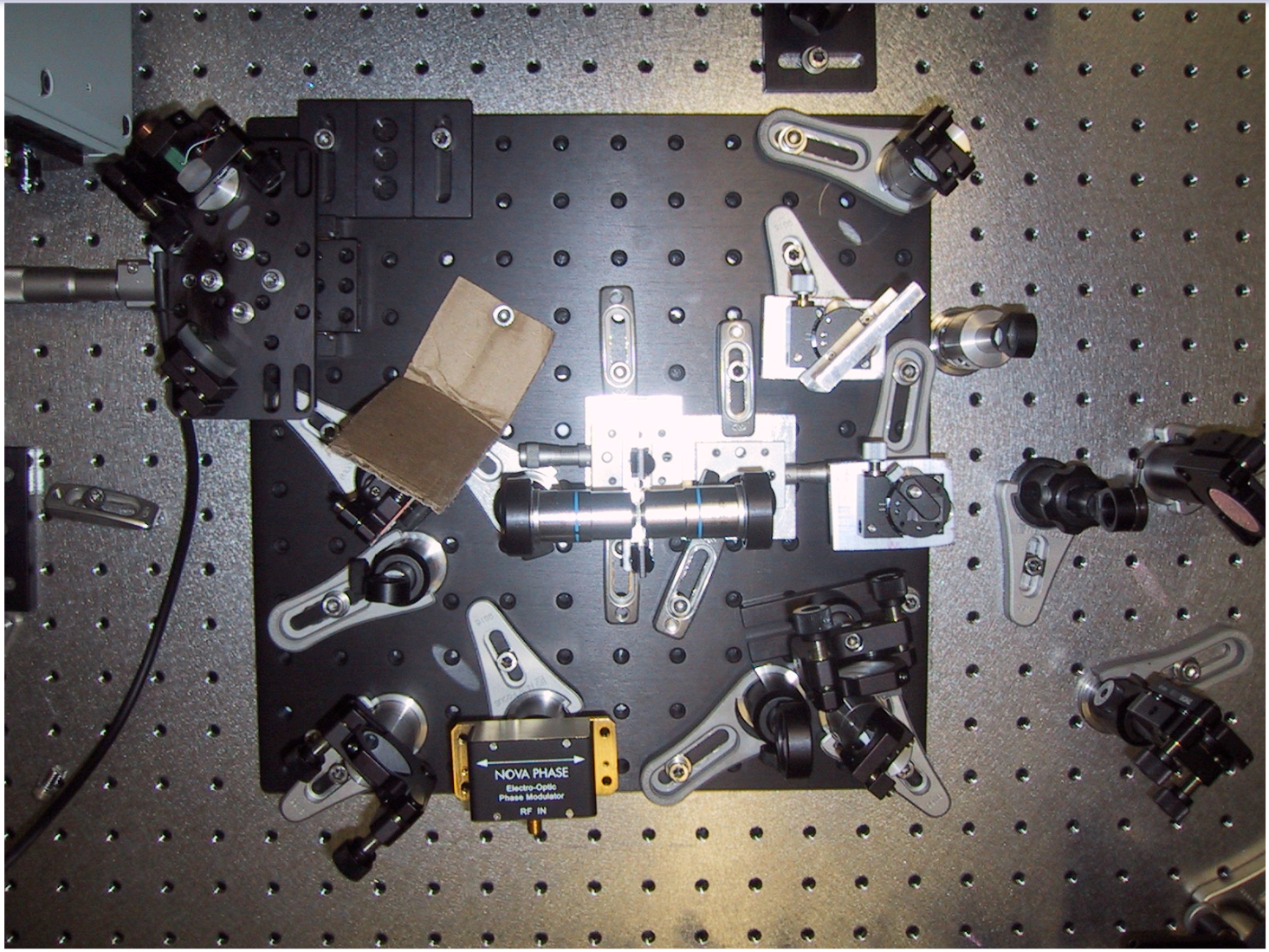

DMSO Interference

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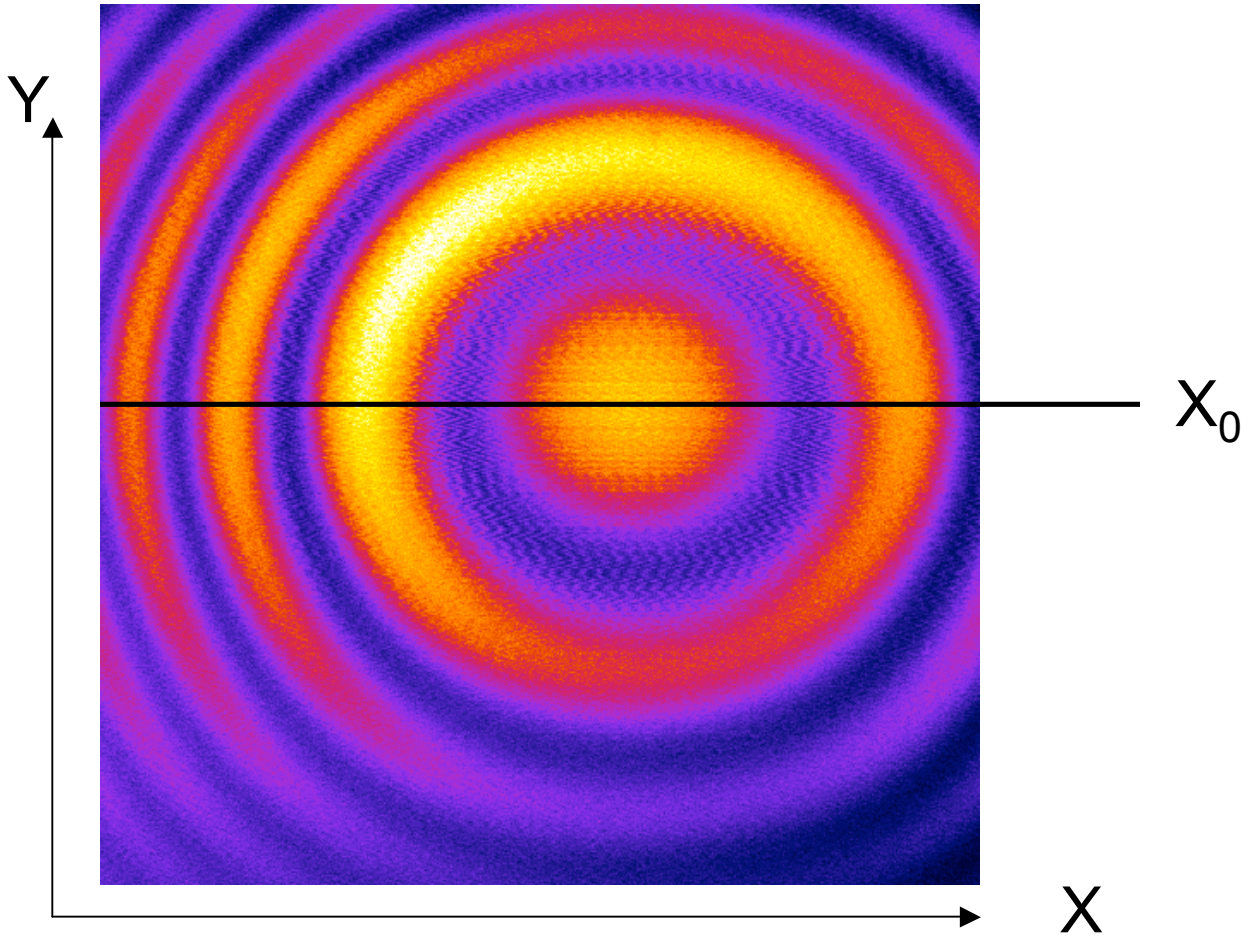
Interference between local oscillator and CARS signal



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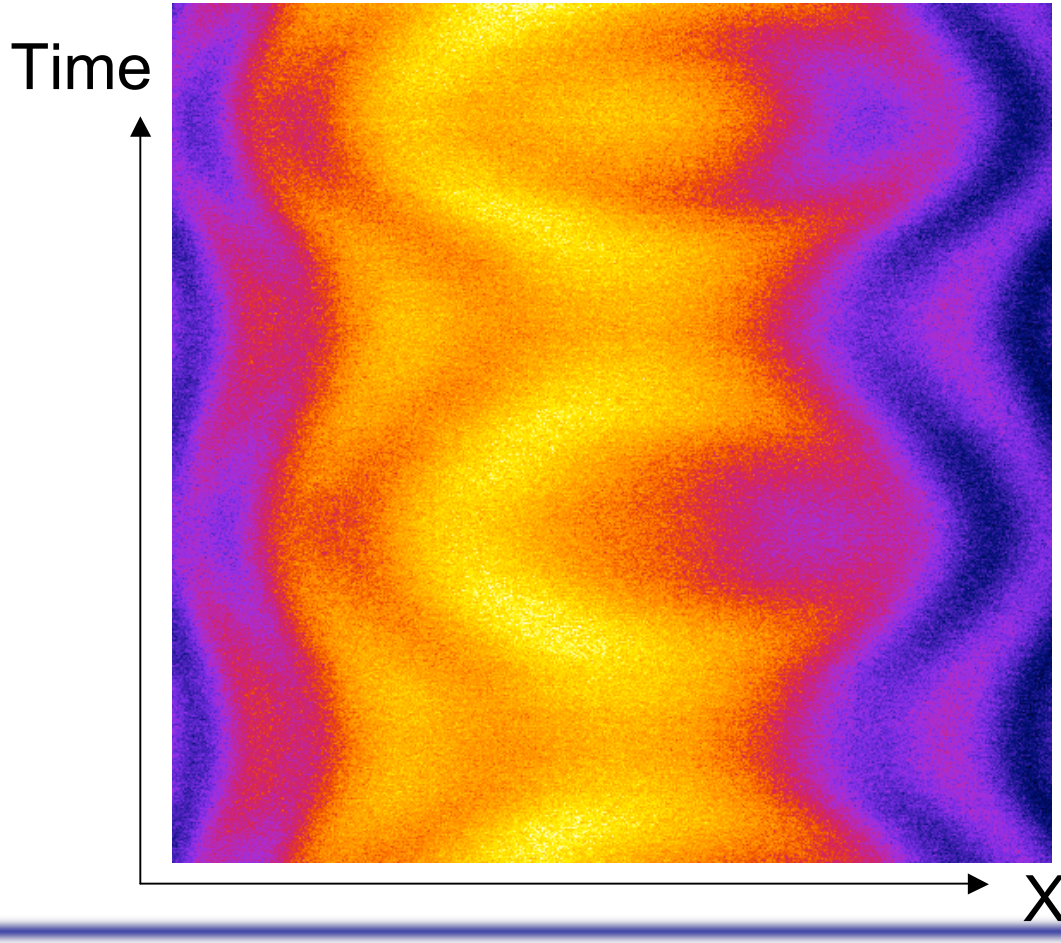

DMSO-Glass Experiment

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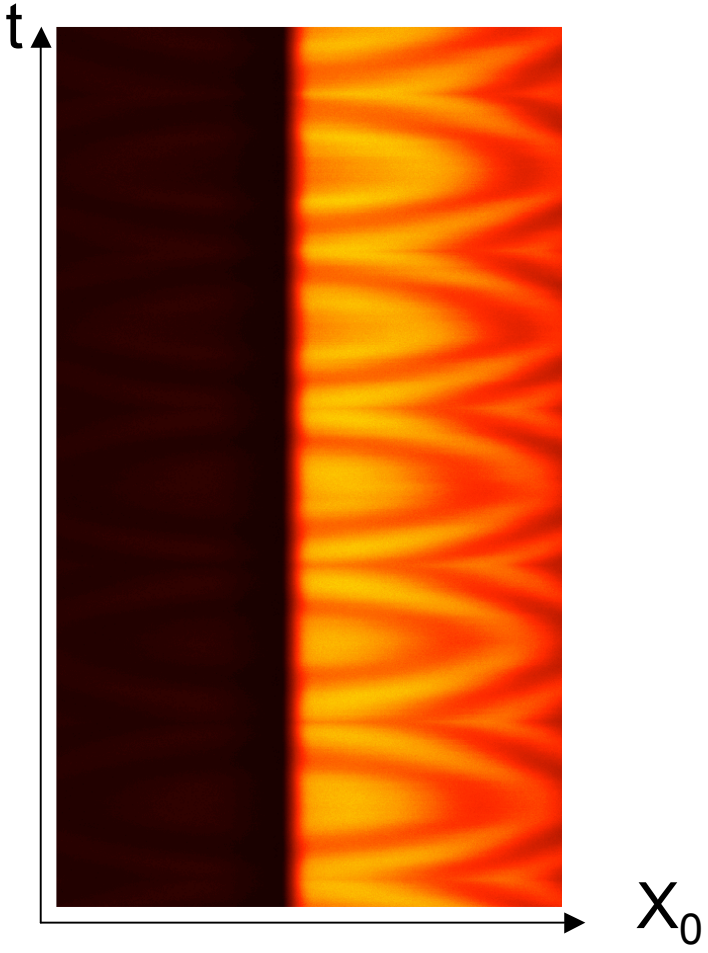
Simultaneous observation of two heterodyne signals

+

local oscillator



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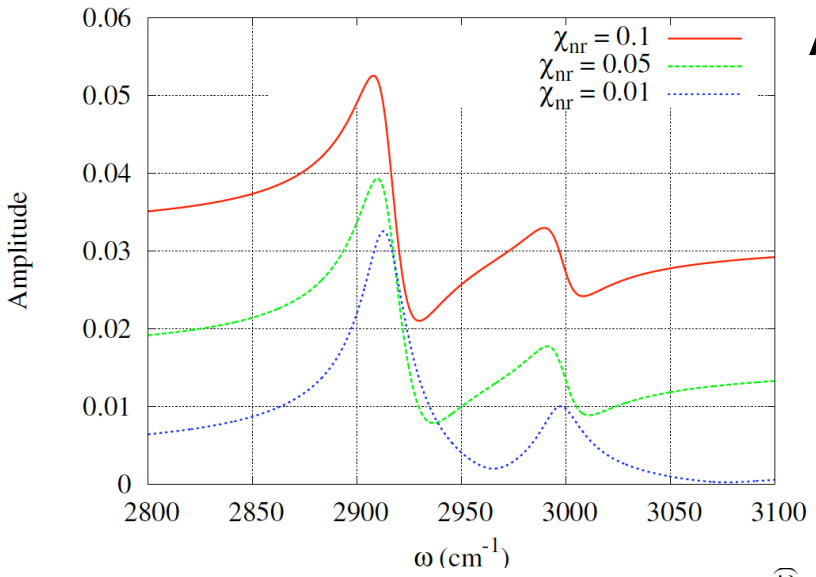
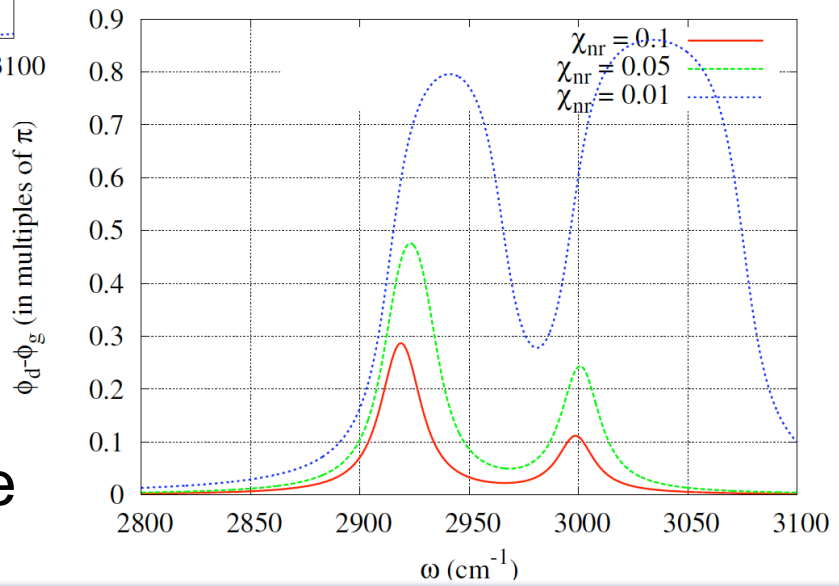


Amplitude of oscillation

Phase difference



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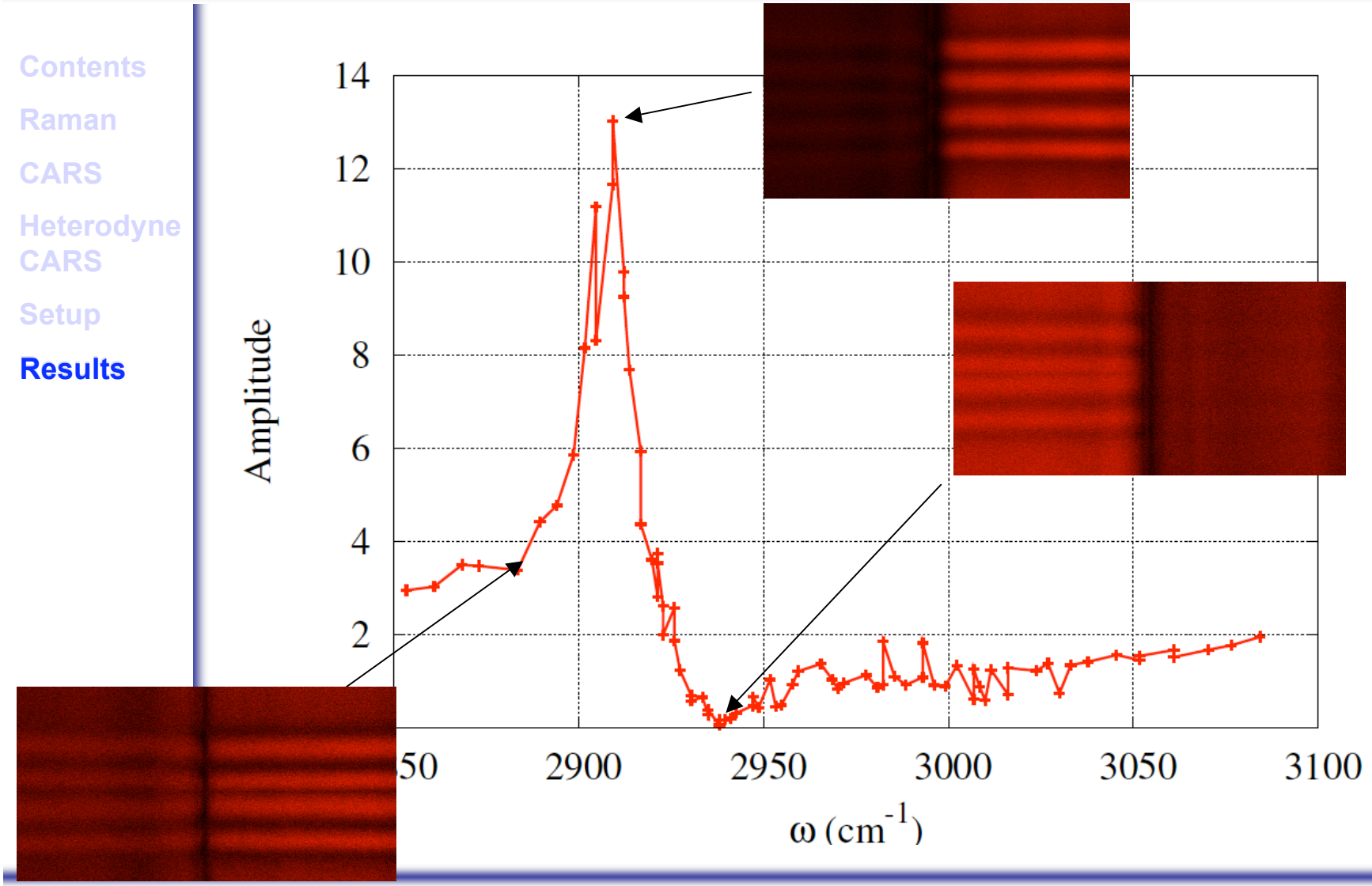

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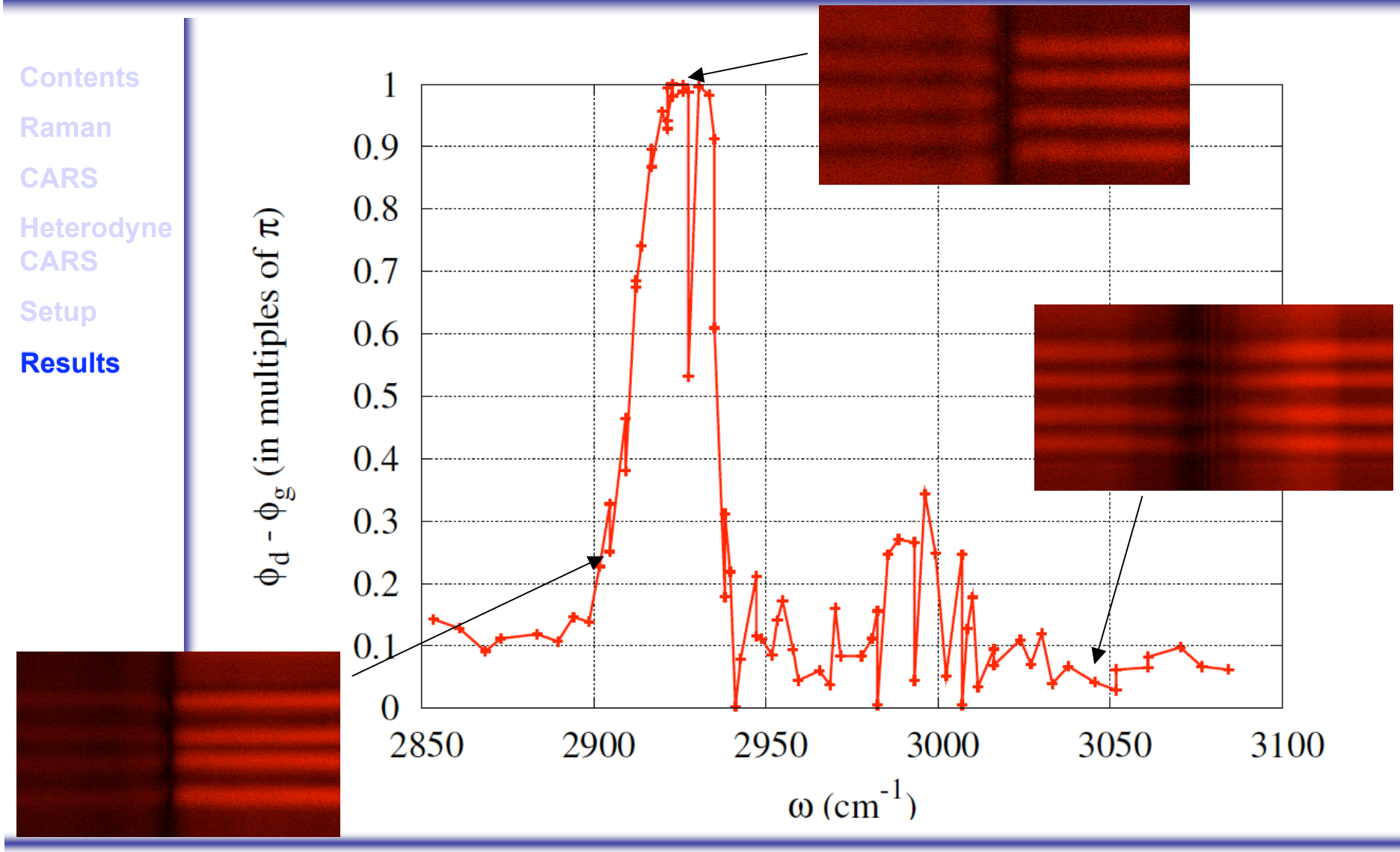

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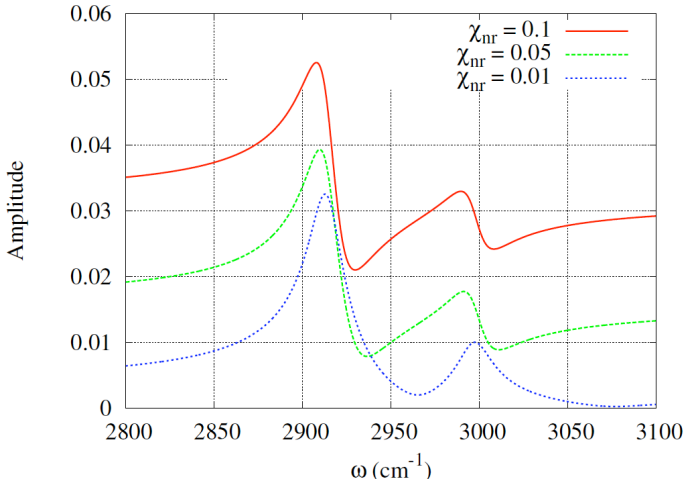

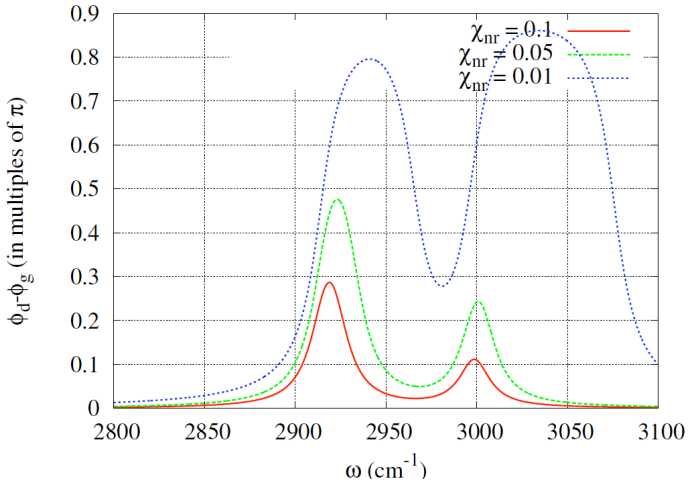

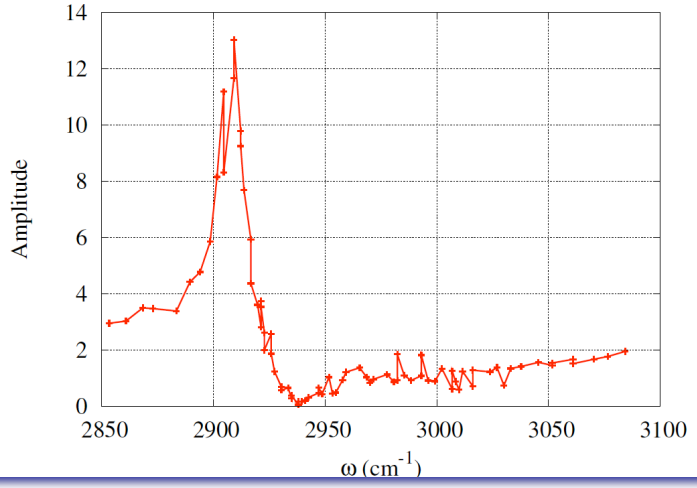

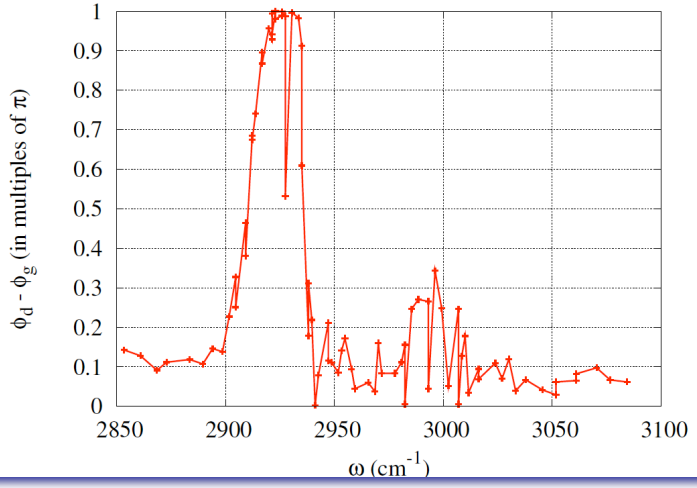

Conclusion

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- Raman versus CARS
  - Non-resonant background and its role
  - CARS intensity is not proportional to conc.
- CARS Heterodyne setup
- Experiments with DMSO + Glass