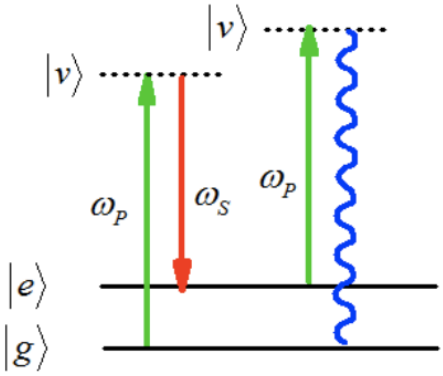
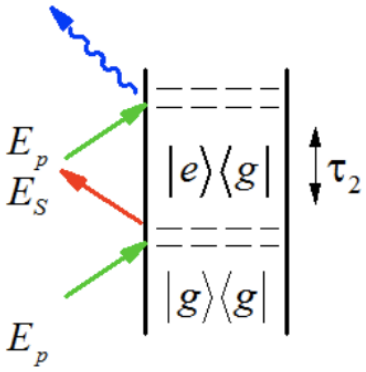
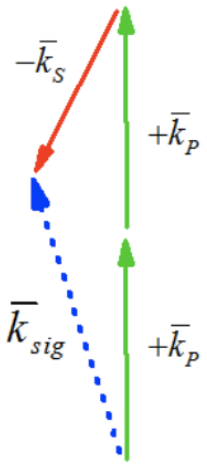
3.5: CARS (Coherent Anti-Stoke Raman Scattering)

Used to drive ground state vibrations with optical pulses or cw fields.

- Two fields, with a frequency difference equal to a vibrational transition energy, are used to excite the vibration.
- The first field is the “pump” and the second is the “Stokes” field.
- A second interaction with the pump frequency lead to a signal that radiates at the anti-Stokes frequency: $\omega_{sig} = 2\omega_P - \omega_S$ and the signal is observed background-free next to the transmitted pump field: $\bar{k}_{sig} = 2\bar{k}_P - \bar{k}_S$.







The experiment is described by R_1 to R_4, and the polarization is

$$\begin{aligned} R^{(3)} &= \bar{\mu}_{ev'}\bar{\mu}_{v'g}e^{-i\omega_{eg}\tau-\Gamma_{eg}\tau}\bar{\mu}_{gv}\bar{\mu}_{ve} + c.c. \\ &= \bar{\alpha}_{eg}e^{-i\omega_{eg}\tau-\Gamma_{eg}\tau}\bar{\alpha}_{ge} + c.c. \end{aligned}$$

The CARS experiment is similar to a linear experiment in which the lineshape is determined by the Fourier transform of $C(\tau) = \langle\bar{\alpha}(\tau)\bar{\alpha}(0)\rangle$.

The same processes contribute to Optical Kerr Effect Experiments and Impulsive Stimulated Raman Scattering.