• In NMR, signals result from a series of $\pi/2$ or $\pi$ pulses. Wavelengths are very long compared to sample.

• In optical analogues, signals result created by coherent, phased array of oscillators created by interaction with 3 excitation fields. Wavelengths are very short compared to sample.

• Momentum conservation requires $\vec{k}_4 = \vec{k}_1 - \vec{k}_2 + \vec{k}_3$

• Signal vanishes if coherence dephases so dephasing rates define the time scale of the spectral measurement.

• Three interactions allows one to follow dynamics

• Frequency or time domain measurements
Single quantum coherence:
\[ \Psi(x,t) = c_a(t) \psi_a(x) e^{i\omega_a t} + c_b(t) \psi_b(x) e^{i\omega_b t} \]

Coherences are time dependent:
\[
|\Psi(x,t)|^2 = |c_a(t)|^2 |\psi_a(x)|^2 + |c_b(t)|^2 |\psi_b(x)|^2 \\
+ c_a(t)c^*_b(t) \psi_a \psi_b^* e^{i(\omega_a - \omega_b)t} + c^*_a(t)c_b(t) \psi_a^* \psi_b e^{-i(\omega_a - \omega_b)t}
\]

Density matrices define coherences
\[
|\psi_b \rangle \langle \psi_a | \Rightarrow c_b c^*_a
\]
Absorption, Refraction

Sum & Difference Frequency Generation
Heterodyned Stimulated Photon Echo

\[ v'' = v + v' \]

\[ 2v' \]

\[ 2v \]

\[ v' \]

\[ v \]

\[ g \]

\[ v' \]

\[ v \]

\[ g \]

\[ v'' = v + v' \]

\[ 2v' \]

\[ 2v \]

\[ v' \]

\[ v \]

\[ g \]

\[ v' \]

\[ v \]

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\[ v \]

\[ g \]