Nonlinear Optics

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Nonlinear Susceptibilities

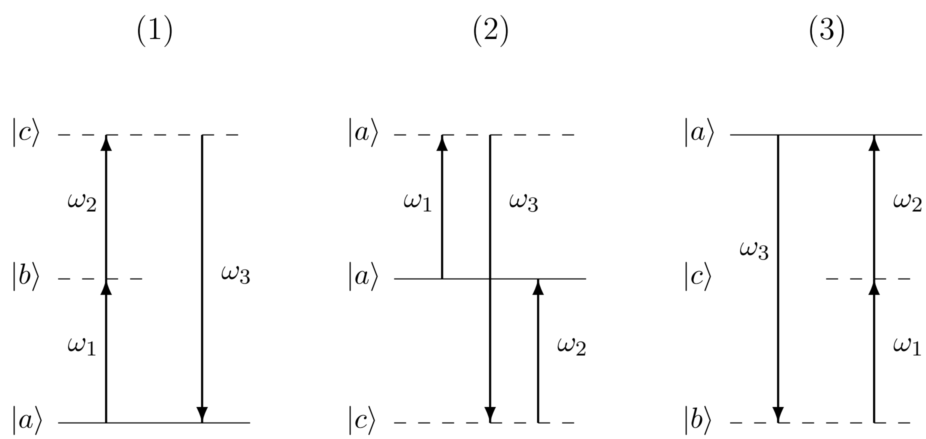
Coherent nonlinear phenomena are based upon the scattering amplitudes $U_a a$ because the final state must be the initial state. A coherent interaction requires summation of all the scattering amplitudes through a macroscopic volume because the relative phases are the quantities that lead to coherence.

Second-Order Susceptibilities

In an unfortunate display of poor vocabulary, a coherent process which involves three photons is referred to as a second-order process governed by a second-order susceptibility tensor $\chi^{(2)}$. The nanoscopic version is called β, and, for the case $\omega_3 = \omega_1 + \omega_2$, it will be approximated by analogy to the two-photon scattering tensor as

$$\beta = \sum_{b,c} \left[\frac{\langle a|r_3|c\rangle\langle c|r_2|b\rangle\langle b|r_1|a\rangle}{(\omega_{ba} - \omega_1)(\omega_{ca} - (\omega_1 + \omega_2))} + \frac{\langle a|r_2|c\rangle\langle c|r_3|b\rangle\langle b|r_1|a\rangle}{(\omega_{ba} - \omega_1)(\omega_{ca} - (\omega_1 - \omega_3))} + \frac{\langle a|r_2|c\rangle\langle c|r_1|b\rangle\langle b|r_3|a\rangle}{(\omega_{ba} + \omega_3)(\omega_{ca} - (\omega_1 - \omega_3))} \right] . \tag{1}$$

The three time-orderings are depicted below.



Note that if $\omega_1 \neq \omega_2$ then there should be six time-orderings. The applied field amplitudes are E_1 and E_2. The scattering amplitude is defined, within a few important scalar factors, in terms of the tensor of rank three and the applied fields as

$$U_{aa} = e^{i(\vec{k}_1 + \vec{k}_2 - \vec{k}_3)\cdot\vec{r}_\circ} E_1 E_2 \beta \hat{\epsilon}_1 \hat{\epsilon}_2 = \beta \vec{E}_1 \vec{E}_2 e^{i\Delta\vec{k}\cdot\vec{r}_\circ} . \tag{2}$$

Third-Order Susceptibilities

A coherent process which involves four photons is referred to as a third-order process and is governed by a third-order susceptibility tensor $\chi^{(3)}$. The nanoscopic version is called γ, and, for the case $\omega_4 = \omega_1 \pm \omega_2 \pm \omega_3$, is the fourth-rank tensor

$$\gamma = \sum_{b,c,d} \frac{\langle a|r_4|d\rangle\langle d|r_3|c\rangle\langle c|r_2|b\rangle\langle b|r_1|a\rangle}{(\omega_{ba}-\omega_1)(\omega_{ca}-(\omega_1\pm\omega_2))(\omega_{da}-(\omega_1\pm\omega_2\pm\omega_3))} \tag{3}$$

$$+\frac{\langle a|r_3|d\rangle\langle d|r_4|c\rangle\langle c|r_2|b\rangle\langle b|r_1|a\rangle}{(\omega_{ba}-\omega_1)(\omega_{ca}-(\omega_1\pm\omega_2))(\omega_{da}-(\omega_1\pm\omega_2\omega_4))}$$

$$+\frac{\langle a|r_3|d\rangle\langle d|r_2|c\rangle\langle c|r_4|b\rangle\langle b|r_1|a\rangle}{(\omega_{ba}-\omega_1)(\omega_{ca}-(\omega_1\omega_4))(\omega_{da}-(\omega_1-\omega_4\pm\omega_2}$$

$) + \cdots .$

Note that if if all the photons are of different energies, then there should be 24 time-orderings. The applied field amplitudes are E_1, E_2 and E_3. The scattering amplitude is defined, within a few important scalar factors, in terms of the tensor of rank four and the applied fields as

$$U_{aa} = \gamma\vec{E}_1\vec{E}_2\vec{E}_3 e^{i(\vec{k}_1\pm\vec{k}_2\pm\vec{k}_3-\vec{k}_4)\cdot\vec{r}_\circ} = \gamma\vec{E}_1\vec{E}_2\vec{E}_3 e^{i\Delta\vec{k}\cdot\vec{r}_\circ} . \tag{4}$$