Microscopic Cascading in Fifth-Order Nonlinearity Induced by Local-Field Effects

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Cascading

In a broad sense:

\( \chi_{\text{eff}}^{(3)} = \text{const} \times \chi^{(2)} : \chi^{(2)} \)
Cascading

\[ \gamma^{(2)} = 2\omega \]

\[ \omega \quad + \quad \omega \quad = \quad 2\omega \]

\[ L_{\text{coh, \, 1}} \]
Cascading

\[ \omega + \omega = \chi^{(2)} = 2 \omega \]

\[ 2 \omega + \omega = \chi^{(2)} = 3 \omega \]

\[ L_{\text{coh}, 1} \]

\[ L_{\text{coh}, 2} \]
Cascading

Macroscopic: requires propagation and phase-matching
Cascading

\[ \omega + \omega = 2\omega \]

Macroscopic: requires propagation and phase-matching
Due to local-field effects
Cascading

\[
\omega + \chi^{(2)} + \omega = 2\omega + \chi^{(2)} + \text{LFE} + \omega = 3\omega
\]

Due to local-field effects

**Microscopic**: does not require propagation and phase-matching
Consider a homogeneous medium exposed to an external optical field:

$E_{\text{ext}}$
Consider a homogeneous medium exposed to an external optical field:

\[ E_{\text{ext}} \neq E_{\text{loc}} \neq E \]
Lorentz Local Field

\[ E_{\text{loc}} \neq E \]

Imaginary sphere (boundary of virtual cavity)

Contributions from inside dipoles are accounted exactly

The dipoles outside the cavity are considered as a homogeneous medium

\[ b \ll R \ll \lambda \]
Lorentz Local Field

\[ E_{\text{loc}} \neq E \]

- \( b \ll R \ll \lambda \)
- \( E \) is average (macroscopic) field in the medium
- \( E_{\text{loc}} \) is the local field acting on a typical emitter
- \( P \) is average (macroscopic) polarization

\[ E_{\text{loc}} = E + \frac{4\pi}{3} P \]

Imaginary sphere (boundary of virtual cavity)
Contributions from inside dipoles are accounted exactly
The dipoles outside the cavity are considered as a homogeneous medium
Lorentz Local Field

\[ E_{\text{loc}} = E + \frac{4\pi}{3} P \quad \text{or} \quad E_{\text{loc}} = L E \]
Lorentz Local Field

\[ E_{\text{loc}} = E + \frac{4\pi}{3} P \]

or

\[ E_{\text{loc}} = L E \]

where

\[ L = \frac{\epsilon^{(1)} + 2}{3} \]

is Lorentz local-field correction factor

\[ \epsilon^{(1)} \]

is dielectric permittivity
Two-Level Atom

\[ \Delta = \omega - \omega_{ba} \]

\[ \tilde{E} = E(t) \exp(-i \omega t) \]

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Two-Level Atom

\[ \Delta = \omega - \omega_{ba} \]

\[ \tilde{E} = E(t) \exp(-i \omega t) \]

\[ \hbar \omega_{ba} \]

\[ \frac{1}{T_1} \]

\[ \frac{2}{T_2} \]
A Collection of Two-Level Atoms
Maxwell-Bloch Equations

\[ \dot{\sigma} = \left( i \Delta - \frac{1}{T_2} \right) \sigma - \frac{1}{2} i \kappa Ew \]

\[ \dot{w} = -\frac{w - w^{eq}}{T_1} + i \left( \kappa E \sigma^* - \kappa^* E^* \sigma \right) \]

- \( \sigma \) is coherence
- \( w \) is population inversion
- \( w^{eq} \) is equilibrium population inversion
- \( \kappa = 2 \mu / \hbar \) is atom-field coupling constant
- \( \Delta \) is detuning
- \( \mu \) is transition dipole moment
- \( T_1 \) is population relaxation time
- \( T_2 \) is coherence relaxation time
Maxwell-Bloch Equations

\[ \dot{\sigma} = \left( i \Delta - \frac{1}{T_2} \right) \sigma - \frac{1}{2} i \kappa E_{\text{loc}} \]

\[ \dot{\omega} = - \frac{\omega - \omega^{\text{eq}}}{T_1} + i \left( \kappa E_{\text{loc}} \sigma^* - \kappa^* E_{\text{loc}}^* \sigma \right) \]

\[ E_{\text{loc}} = E + \frac{4 \pi}{3} P \]
Steady-State Solutions

\[ w = -\frac{1}{1 + \frac{|E|^2 / |E_s|^2}{1 + T_2^2(\Delta + \Delta_L w)^2}} \]

\[ \sigma = \frac{\mu}{\hbar} \frac{wE}{\Delta + \Delta_L w + i/T_2} \]
Steady-State Solutions

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\[ \sigma = \frac{\mu}{\hbar} \frac{wE}{\Delta + \Delta_L w + i/T_2} \]

\[ \Delta_L = -\frac{4\pi N|\mu|^2}{3\hbar} \]

inversion-dependent frequency shift

Lorentz red shift
Polarization

\[ P = N \mu^* \sigma \]
Polarization

\[ P = N \mu^* \sigma = \chi E \]
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Local-field-corrected

\[ \sigma = \frac{\mu}{\hbar} \frac{wE}{\Delta + \Delta L w + i/T_2} \]
Polarization

\[ P = N \mu^* \sigma = \chi E \]

Local-field-corrected

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Susceptibilities

\[ P = \chi E = \chi^{(1)} E + 3 \chi^{(3)} |E|^2 E + 10 \chi^{(5)} |E|^4 E \]
Susceptibilities

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Susceptibilities

Local-field-corrected

\[ P = \chi E = \chi^{(1)} E + 3 \chi^{(3)} |E|^2 E + 10 \chi^{(5)} |E|^4 E \]
LF-Corrected Linear and Nonlinear Susceptibilities

The result:

\[ \chi^{(1)} = N \gamma^{(1)}_{at} L; \]

\[ \chi^{(3)} = N \gamma^{(3)}_{at} |L|^2 L^2; \]

\[ \chi^{(5)} = N \gamma^{(5)}_{at} |L|^4 L^2 \]
\[ + \frac{24 \pi}{10} N^2 (\gamma^{(3)}_{at})^2 |L|^4 L^3 + \frac{12 \pi}{10} N^2 |\gamma^{(3)}_{at}|^2 |L|^6 L. \]

\( \gamma^{(1)}_{at} \) is microscopic polarizability

\( \gamma^{(3)}_{at} \) and \( \gamma^{(5)}_{at} \) are 3\textsuperscript{rd}- and 5\textsuperscript{th}-order hyperpolarizabilities
The result:

well-known

\[ \chi^{(1)} = N \gamma^{(1)}_{at} L ; \]
\[ \chi^{(3)} = N \gamma^{(3)}_{at} |L|^2 L^2 ; \]
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The result:

well-known \[ \chi^{(1)} = N \gamma_{at}^{(1)} L ; \]
\[ \chi^{(3)} = N \gamma_{at}^{(3)} |L|^2 L^2 ; \]
\[ \chi^{(5)} = N \gamma_{at}^{(5)} |L|^4 L^2 \]
\[ + \frac{24\pi}{10} N^2 (\gamma_{at}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma_{at}^{(3)}|^2 |L|^6 L . \]
The result:

well-known

\[ \chi^{(1)} = N \gamma^{(1)}_{at} L; \]

nothing peculiar

\[ \chi^{(3)} = N \gamma^{(3)}_{at} |L|^2 L^2; \]

\[ \chi^{(5)} = N \gamma^{(5)}_{at} |L|^4 L^2 \]

\[ + \frac{24 \pi}{10} N^2 (\gamma^{(3)}_{at})^2 |L|^4 L^3 + \frac{12 \pi}{10} N^2 |\gamma^{(3)}_{at}|^2 |L|^6 L. \]
The result:

well-known $\chi^{(1)} = N \gamma^{(1)}_{at} L$;

nothing peculiar $\chi^{(3)} = N \gamma^{(3)}_{at} |L|^2 L^2$;

peculiar $\chi^{(5)} = N \gamma^{(5)}_{at} |L|^4 L^2$

$$+ \frac{24 \pi}{10} N^2 (\gamma^{(3)}_{at})^2 |L|^4 L^3 + \frac{12 \pi}{10} N^2 |\gamma^{(3)}_{at}|^2 |L|^6 L.$$
The result:

\[ \chi^{(1)} = N \gamma^{(1)}_{\text{at}} L ; \]

\[ \chi^{(3)} = N \gamma^{(3)}_{\text{at}} |L|^2 L^2 ; \]

\[ \chi^{(5)} = N \gamma^{(5)}_{\text{at}} |L|^4 L^2 + \frac{24\pi}{10} N^2 (\gamma^{(3)}_{\text{at}})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma^{(3)}_{\text{at}}|^2 |L|^6 L . \]
LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

$$\chi^{(5)} = N \gamma_{at}^{(5)} |L|^4 L^2$$

$$+ \frac{24 \pi}{10} N^2 (\gamma_{at}^{(3)})^2 |L|^4 L^3 + \frac{12 \pi}{10} N^2 |\gamma_{at}^{(3)}|^2 |L|^6 L.$$
"direct" contribution from fifth-order hyperpolarizability $\gamma^{(5)}_{at}$

$$\chi^{(5)} = N \gamma^{(5)}_{at} |L|^4 L^2$$
$$+ \frac{24 \pi}{10} N^2 (\gamma^{(3)}_{at})^2 |L|^4 L^3 + \frac{12 \pi}{10} N^2 |\gamma^{(3)}_{at}|^2 |L|^6 L.$$
LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

"direct" contribution from fifth-order hyperpolarizability $\gamma^{(5)}_{at}$

$$\chi^{(5)} = N \gamma^{(5)}_{at} |L|^4 L^2$$

$$+ \frac{24 \pi}{10} N^2 \left( \gamma^{(3)}_{at} \right)^2 |L|^4 L^3$$

$$+ \frac{12 \pi}{10} N^2 \left| \gamma^{(3)}_{at} \right|^2 |L|^6 L.$$
LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

$\chi^{(5)}_{\text{direct}} = N \gamma_{\text{at}}^{(5)} |L|^4 L^2$ scales as 6th power of factor $L$.

$\chi^{(5)}_{\text{cascaded}} = \frac{24 \pi}{10} N^2 (\gamma_{\text{at}}^{(3)})^2 |L|^4 L^3 + \frac{12 \pi}{10} N^2 |\gamma_{\text{at}}^{(3)}|^2 |L|^6 L$ scales as 7th power of factor $L$. 
LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

$$\chi^{(5)}_{\text{direct}} = N \gamma^{(5)}_{\text{at}} |L|^4 L^2$$
scales as 6\textsuperscript{th} power of factor $L$.

$$\chi^{(5)}_{\text{cascaded}} = \frac{24 \pi}{10} N^2 (\gamma^{(3)}_{\text{at}})^2 |L|^4 L^3$$
scales as 7\textsuperscript{th} power of factor $L$.

How significant?
Consider sodium $3s \rightarrow 3p$ transition:

- The dipole moment $|\mu| = 5.5 \times 10^{-18}$ esu
- Population relaxation time $T_1 = 16$ ns
- Atomic density range $N = 10^{13} - 10^{17}$ cm$^{-3}$
Direct and Cascaded Contributions: Comparison

\[ N = 1 \times 10^{13} \text{ cm}^{-3} \]

\[ \chi^{(5)} \]

\[ \Delta T_2 \]

- Re(\(\chi_{\text{direct}}^{(5)}\))
- Im(\(\chi_{\text{direct}}^{(5)}\))
- Re(\(\chi^{(5)}\))
- Im(\(\chi^{(5)}\))
Direct and Cascaded Contributions: Comparison

N = 1 \times 10^{14} \text{ cm}^{-3}

\Delta T_2
Direct and Cascaded Contributions: Comparison

\[ N = 1 \times 10^{16} \text{ cm}^{-3} \]

\[ \chi^{(5)} \]

\[ \Delta T_2 \]

Phases:
- \( \text{Re}(\chi^{(5)}_{\text{direct}}) \)
- \( \text{Im}(\chi^{(5)}_{\text{direct}}) \)
- \( \text{Re}(\chi^{(5)}) \)
- \( \text{Im}(\chi^{(5)}) \)
Direct and Cascaded Contributions: Comparison

$N = 1 \times 10^{17} \text{ cm}^{-3}$
Direct and Cascaded Contributions: Comparison

N = $1 \times 10^{13}$ cm$^{-3}$

$\chi(5)$

$\Delta T_2$

N = $1 \times 10^{14}$ cm$^{-3}$

$\chi(6)$

$\Delta T_2$

N = $1 \times 10^{16}$ cm$^{-3}$

N = $1 \times 10^{17}$ cm$^{-3}$
Direct and Cascaded Contributions: Comparison

Ratio of absolute values of the contributions as a function of the normalized detuning and atomic density

$$R = \left| \frac{\chi^{(5)}_{\text{cascaded}}}{\chi^{(5)}_{\text{direct}}} \right|$$
Under certain conditions, the cascaded contribution can be as large as the direct contribution.

\[ R = \frac{|\chi_{\text{cascaded}}^{(5)}|}{|\chi_{\text{direct}}^{(5)}|} \]
Microscopic cascading is possible due to local-field-induced contributions of lower-order nonlinearities to higher-order nonlinearities.

We demonstrated it based on Maxwell-Bloch equations for a collection of two-level atoms.

We demonstrated that the cascaded contribution to $\chi^{(5)}$ can be as large as the direct contribution.

Experiment is in progress to verify the theory.
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