Cascade-Like Nonlinearity Caused by Local-Field Effects: Extending Bloembergen's Result

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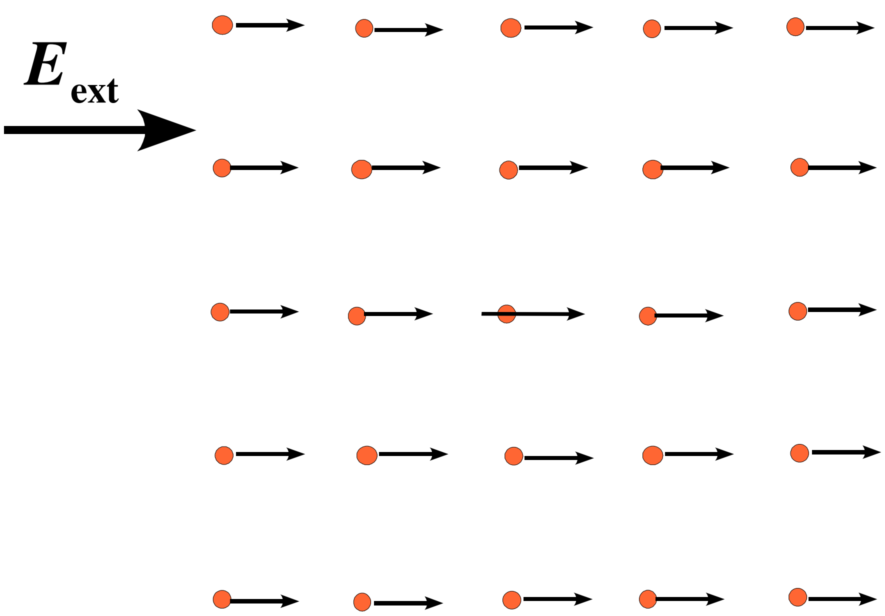
Motivation

- Cascading is a nonlinear process in which a lower-order nonlinearity contributes to a higher-order nonlinear response in a multi-step fashion: $\chi^{(3)} = \text{const} \times \chi^{(2)} : \chi^{(2)}$.
- Cascading in a usual sense requires propagation and phase-matching.
- Local-field effects can cause cascading at a microscopic level. Such cascading does not require propagation and phase matching.
- Although macroscopic cascading is a well-known phenomenon, microscopic cascading is less well known.

Lorentz Local Field

Consider a homogeneous medium exposed to an external optical field:



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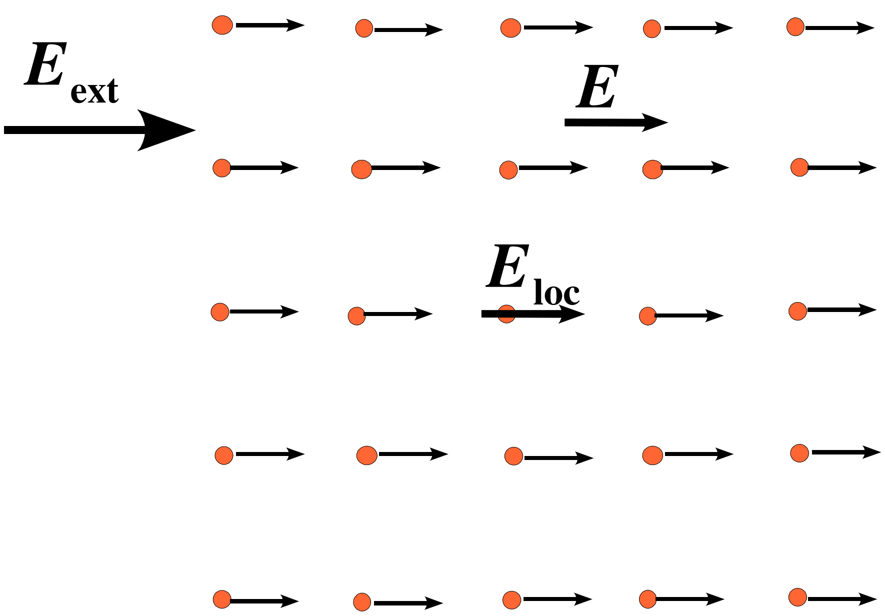
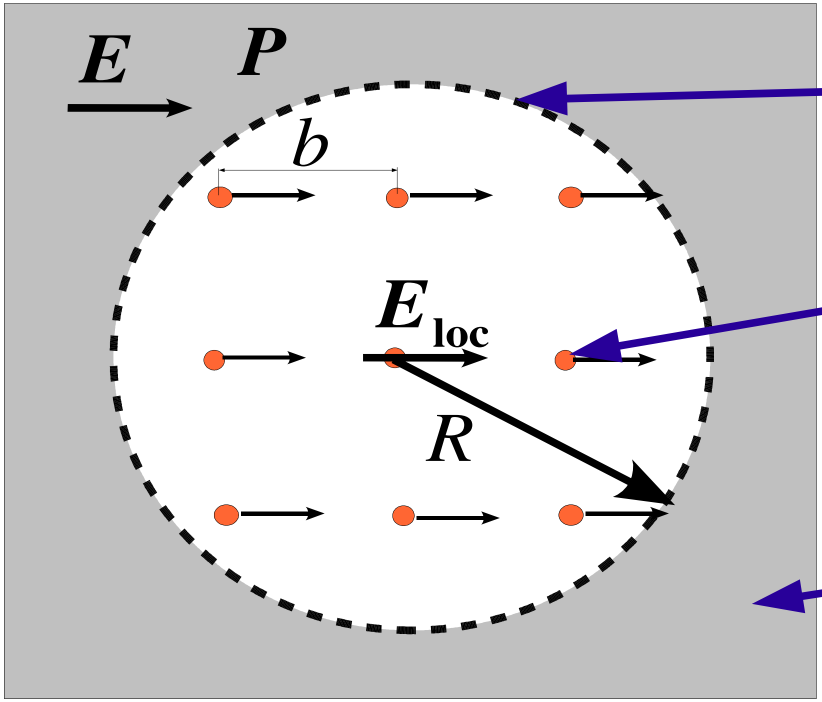


$$E_{loc} \neq E_{ext} \neq E$$

Lorentz Local Field

$$\boldsymbol{E}_{\text{loc}} \neq \boldsymbol{E}$$



Imaginary sphere (boundary of virtual cavity)

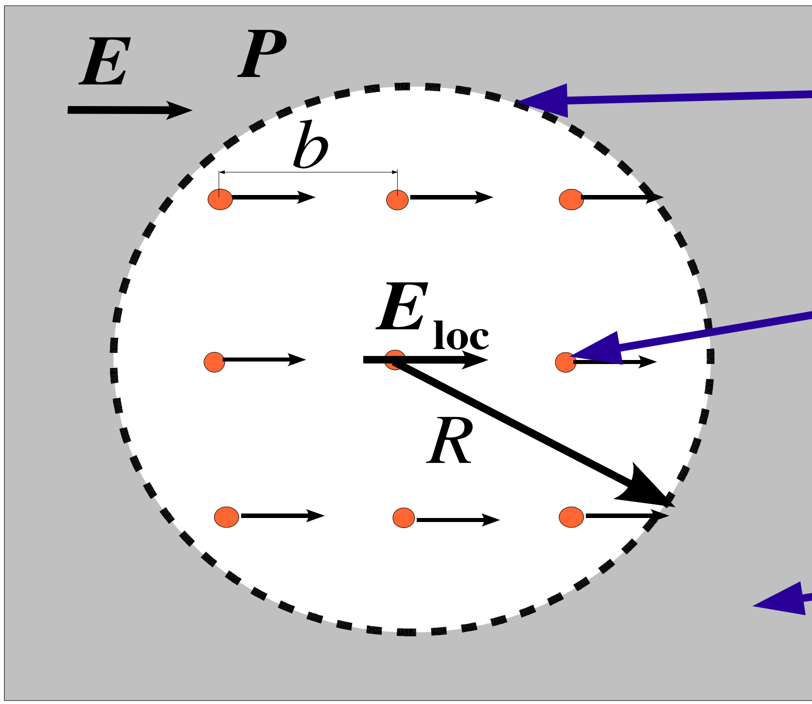
Contributions from inside dipoles are accounted exactly

The dipoles outside the cavity are considered as a homogeneous medium

$$b \ll R \ll \lambda$$

Lorentz Local Field

$$\boldsymbol{E}_{\text{loc}} \neq \boldsymbol{E}$$



Imaginary sphere (boundary of virtual cavity)

Contributions from inside dipoles are accounted exactly

The dipoles outside the cavity are considered as a homogeneous medium

$$b \ll R \ll \lambda$$

$$\boldsymbol{E}_{\text{loc}} = \boldsymbol{E} + \frac{4\pi}{3}\boldsymbol{P}$$

$\boldsymbol{E}$ is average (macroscopic) field in the medium

$\boldsymbol{E}_{\text{loc}}$ is the local field acting on a typical emitter

$\boldsymbol{P}$ is average (macroscopic) polarization

Lorentz Local Field

$$\mathbf{E}_{\mathrm{loc}} = \mathbf{E} + \frac{4\pi}{3}\mathbf{P}$$

or

$$\mathbf{E}_{\mathrm{loc}} = L\,\mathbf{E}$$

Lorentz Local Field

$$\boldsymbol{E}_{\mathrm{loc}} = \boldsymbol{E} + \frac{4\pi}{3}\boldsymbol{P}$$

or

$$\boldsymbol{E}_{\mathrm{loc}} = L\,\boldsymbol{E}$$

where

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

is Lorentz local-field correction factor

$\epsilon^{(1)}$ is dielectric permittivity

Local-Field Effects in Linear Optics

$$\chi^{(1)} = L\, N\, \gamma_{\text{at}}^{(1)}$$

$\chi^{(1)}$ is linear susceptibility

N is molecular or atomic density

$\gamma_{\text{at}}^{(1)}$ is linear microscopic polarizability

Local-Field Effects in Linear Optics

$$\chi^{(1)} = L\,N\,\gamma_{\text{at}}^{(1)}$$

$\chi^{(1)}$ is linear susceptibility

N is molecular or atomic density

$\gamma_{\text{at}}^{(1)}$ is linear microscopic polarizability

Linear optical properties depend on factor L.

Local-Field Effects in Nonlinear Optics

$$\chi^{(2)}(\omega_3 = \omega_1 + \omega_2) = L_1 L_2 L_3 N \gamma_{\text{at}}^{(2)}$$

where

$$L_i = \frac{\epsilon^{(1)}(\omega_i) + 2}{3}$$

$\chi^{(2)}$ is the second-order nonlinear susceptibility

$\gamma_{\text{at}}^{(2)}$ is the second-order nonlinear hyperpolarizability

N. Bloembergen, *Nonlinear Optics*, 4th ed. (Scientific, Singapore, 1996).

Extending Bloembergen's Result: Naïve Way

Since $\chi^{(2)}$ scales as the 3rd power of factor L,
$\chi^{(n)}$ scales as the $(n+1)^{th}$ power of factor L

Open your favorite textbook on Nonlinear Optics

Extending Bloembergen's Result: Naïve Way

Since $\chi^{(2)}$ scales as the 3rd power of factor L,
$\chi^{(n)}$ scales as the $(n+1)^{th}$ power of factor L

Consider degenerate nth-order nonlinearity:

$$\chi^{(n)}(\omega = \omega - \omega + \omega + \ldots) = N \gamma_{at}^{(n)} |L|^{n-1} L^2$$

Open your favorite textbook on Nonlinear Optics

Third-Order Nonlinearity in Non-Centrosymmetric Media

However,

$$\chi^{(3)}(\omega_3 = 3\omega_1) = \chi_d^{(3)} + \chi_c^{(3)},$$

where

$$\chi_d^{(3)} = N\gamma_{at}^{(3)} L_1^3 L_3$$ and $$\chi_c^{(3)} \propto [\chi^{(2)}]^2 L_2^{-1},$$

$$\chi^{(2)}(\omega_2 = \omega_1 + \omega_1) = N\gamma_{at}^{(2)} L_1^2 L_2 .$$

D. Bedeaux and N. Bloembergen, *Physica* **69**, 57-66 (1973).

Third-Order Nonlinearity in Non-Centrosymmetric Media

$$\chi^{(3)}(\omega_3 = 3\omega_1) = \chi_{\mathrm{d}}^{(3)} + \chi_{\mathrm{c}}^{(3)},$$

where

$\chi_{\mathrm{d}}^{(3)}$ comes from $\gamma_{\mathrm{at}}^{(3)}$ and scales as the 4th power of factor L;

$\chi_{\mathrm{c}}^{(3)}$ comes from $\gamma_{\mathrm{at}}^{(2)}$ and scales as the 5th power of factor L.

D. Bedeaux and N. Bloembergen, *Physica* **69**, 57-66 (1973).

Therefore...

- It appears that $\chi^{(3)}(\omega=\omega+\omega-\omega) \neq N\gamma_{\text{at}}^{(3)}|L|^2 L^2$ in non-centrosymmetric media.
- Straight-forward generalization $\chi^{(n)}(\omega)=N\gamma_{\text{at}}^{(n)}|L|^{n-1}L^2$ of Bloembergen's result for second-order susceptibility to a higher-order susceptibility is, generally, not correct.
- One cannot predict the expression for local-field-corrected $\chi^{(n)}$ based on the result for a lower-order nonlinearity.

Our Contribution

Why the straight-forward generalization does not work?

To answer the question, we undertake calculation of local-field-corrected fifth-order susceptibility in a centrosymmetric medium.

LF-Corrected Degenerate $\chi^{(5)}$: Bloembergen's Prescription

Starting from Lorentz local field

$$E_{\text{loc}} = E + \frac{4\pi}{3} P,$$

$$P = P^{\text{L}} + P^{\text{NL}}$$

consider $P = P^{(1)} + P^{(3)} + P^{(5)},$

where

$$P^{(1)} = N \gamma_{\text{at}}^{(1)} E_{\text{loc}};$$

$$P^{(3)} = N \gamma_{\text{at}}^{(3)} |E_{\text{loc}}|^2 E_{\text{loc}};$$

$$P^{(5)} = N \gamma_{\text{at}}^{(5)} |E_{\text{loc}}|^4 E_{\text{loc}}.$$

centrosymmetric medium

LF-Corrected Degenerate $\chi^{(5)}$: Bloembergen's Prescription

With the use of the nonlinear source polarization

$$P^{\mathrm{NLS}} = L\left(P^{(3)} + P^{(5)}\right),$$

and also $L = \dfrac{\epsilon^{(1)} + 2}{3}$ and $\chi^{(1)} = \dfrac{\epsilon^{(1)} - 1}{4\pi}$,

$$P = P^{(1)} + P^{(3)} + P^{(5)}$$

becomes

$$P = \chi^{(1)} E + P^{\mathrm{NLS}}.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Bloembergen's Prescription

$$P = \chi^{(1)} E + P^{\mathrm{NLS}}$$

Alternatively, as a power-series expansion w. r. t. E:

$$P = \chi^{(1)} E + 3\,\chi^{(3)} |E|^2 E + 10\,\chi^{(5)} |E|^4 E\,.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Bloembergen's Prescription

$$P = \chi^{(1)} E + P^{\mathrm{NLS}}$$

$$P^{\mathrm{NLS}} = L\left(P^{(3)} + P^{(5)}\right)$$

$$P^{(3)} = N \gamma_{\mathrm{at}}^{(3)} \left|E_{\mathrm{loc}}\right|^2 E_{\mathrm{loc}}$$

$$P^{(5)} = N \gamma_{\mathrm{at}}^{(5)} \left|E_{\mathrm{loc}}\right|^4 E_{\mathrm{loc}}$$

Alternatively, as a power-series expansion w. r. t. E:

$$P = \chi^{(1)} E + 3\chi^{(3)} |E|^2 E + 10\chi^{(5)} |E|^4 E .$$

If one can deduce P^{NLS}, one can find $\chi^{(n)}$.

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LF-Corrected Degenerate $\chi^{(5)}$: Wrong Way

$$P = \chi^{(1)} E + P^{\mathrm{NLS}}$$

$$P^{\mathrm{NLS}} = L\left(P^{(3)} + P^{(5)}\right)$$
$$P^{(3)} = N\gamma_{\mathrm{at}}^{(3)} |E_{\mathrm{loc}}|^2 E_{\mathrm{loc}}$$
$$P^{(5)} = N\gamma_{\mathrm{at}}^{(5)} |E_{\mathrm{loc}}|^4 E_{\mathrm{loc}}$$

Substitute *linear* $E_{\mathrm{loc}} = L E$ to get

$$\chi^{(1)} = N\gamma_{\mathrm{at}}^{(1)} L;$$
$$\chi^{(3)} = N\gamma_{\mathrm{at}}^{(3)} |L|^2 L^2;$$
$$\chi^{(5)} = N\gamma_{\mathrm{at}}^{(5)} |L|^4 L^2.$$

Then $\chi^{(n)}(\omega) = N\gamma_{\mathrm{at}}^{(n)} |L|^{n-1} L^2.$

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LF-Corrected Degenerate $\chi^{(5)}$: Correct Way

$$P = \chi^{(1)} E + P^{\text{NLS}}$$

$$P^{\text{NLS}} = L\left(P^{(3)} + P^{(5)}\right)$$

$$P^{(3)} = N\gamma_{\text{at}}^{(3)} \left|E_{\text{loc}}\right|^2 E_{\text{loc}}$$

$$P^{(5)} = N\gamma_{\text{at}}^{(5)} \left|E_{\text{loc}}\right|^4 E_{\text{loc}}$$

Substitute *nonlinear* $E_{\text{loc}} = E + \frac{4\pi}{3} P$

then compare to

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

to deduce $\chi^{(n)}$.

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LF-Corrected Degenerate $\chi^{(5)}$: Correct Result

The result:

$$\chi^{(1)} = N\,\gamma_{\text{at}}^{(1)}\,L\,;$$

$$\chi^{(3)} = N\,\gamma_{\text{at}}^{(3)}\,|L|^2\,L^2\,;$$

$$\chi^{(5)} = N\,\gamma_{\text{at}}^{(5)}\,|L|^4\,L^2 + \frac{24\pi}{10}N^2\left(\gamma_{\text{at}}^{(3)}\right)^2|L|^4\,L^3 + \frac{12\pi}{10}N^2\left|\gamma_{\text{at}}^{(3)}\right|^2|L|^6\,L\,.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Correct Result

The result:

well-known → $\chi^{(1)} = N\gamma_{\text{at}}^{(1)} L;$

$$\chi^{(3)} = N\gamma_{\text{at}}^{(3)} |L|^2 L^2;$$

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

$$+\frac{24\pi}{10} N^2 (\gamma_{\text{at}}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 \left|\gamma_{\text{at}}^{(3)}\right|^2 |L|^6 L.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Correct Result

The result:

well-known → $\chi^{(1)} = N\gamma_{\text{at}}^{(1)} L;$

nothing peculiar → $\chi^{(3)} = N\gamma_{\text{at}}^{(3)} |L|^2 L^2;$

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

$$+\frac{24\pi}{10} N^2 (\gamma_{\text{at}}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma_{\text{at}}^{(3)}|^2 |L|^6 L.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Correct Result

The result:

well-known → $\chi^{(1)} = N\gamma_{\text{at}}^{(1)} L;$

nothing peculiar → $\chi^{(3)} = N\gamma_{\text{at}}^{(3)} |L|^2 L^2;$

deserves attention →

$$\chi^{(5)} = N\gamma_{\text{at}}^{(5)} |L|^4 L^2 + \frac{24\pi}{10} N^2 (\gamma_{\text{at}}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma_{\text{at}}^{(3)}|^2 |L|^6 L.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

$$\chi^{(5)} = N\gamma_{\mathrm{at}}^{(5)}|L|^4 L^2$$
$$+\frac{24\pi}{10}N^2\left(\gamma_{\mathrm{at}}^{(3)}\right)^2|L|^4 L^3 + \frac{12\pi}{10}N^2\left|\gamma_{\mathrm{at}}^{(3)}\right|^2|L|^6 L\,.$$

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LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

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

$$\chi^{(5)} = N \boxed{\gamma_{\text{at}}^{(5)}} |L|^4 L^2 + \frac{24\pi}{10} N^2 (\gamma_{\text{at}}^{(3)})^2 |L|^4 L^3 + \frac{12\pi}{10} N^2 |\gamma_{\text{at}}^{(3)}|^2 |L|^6 L .$$

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LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

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

$$\chi^{(5)} = N \boxed{\gamma_{\text{at}}^{(5)}} |L|^4 L^2 + \frac{24\pi}{10} N^2 \boxed{\left(\gamma_{\text{at}}^{(3)}\right)^2} |L|^4 L^3 + \frac{12\pi}{10} N^2 \boxed{\left|\gamma_{\text{at}}^{(3)}\right|^2} |L|^6 L .$$

"cascaded" contributions from third-order hyperpolarizability $\gamma_{\text{at}}^{(3)}$

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LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

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

scales as 6th power of factor L.

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

scales as 7th power of factor L.

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LF-Corrected Degenerate $\chi^{(5)}$: Direct and Cascaded Contributions

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

scales as 6th power of factor L.

How significant?

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

scales as 7th power of factor L.

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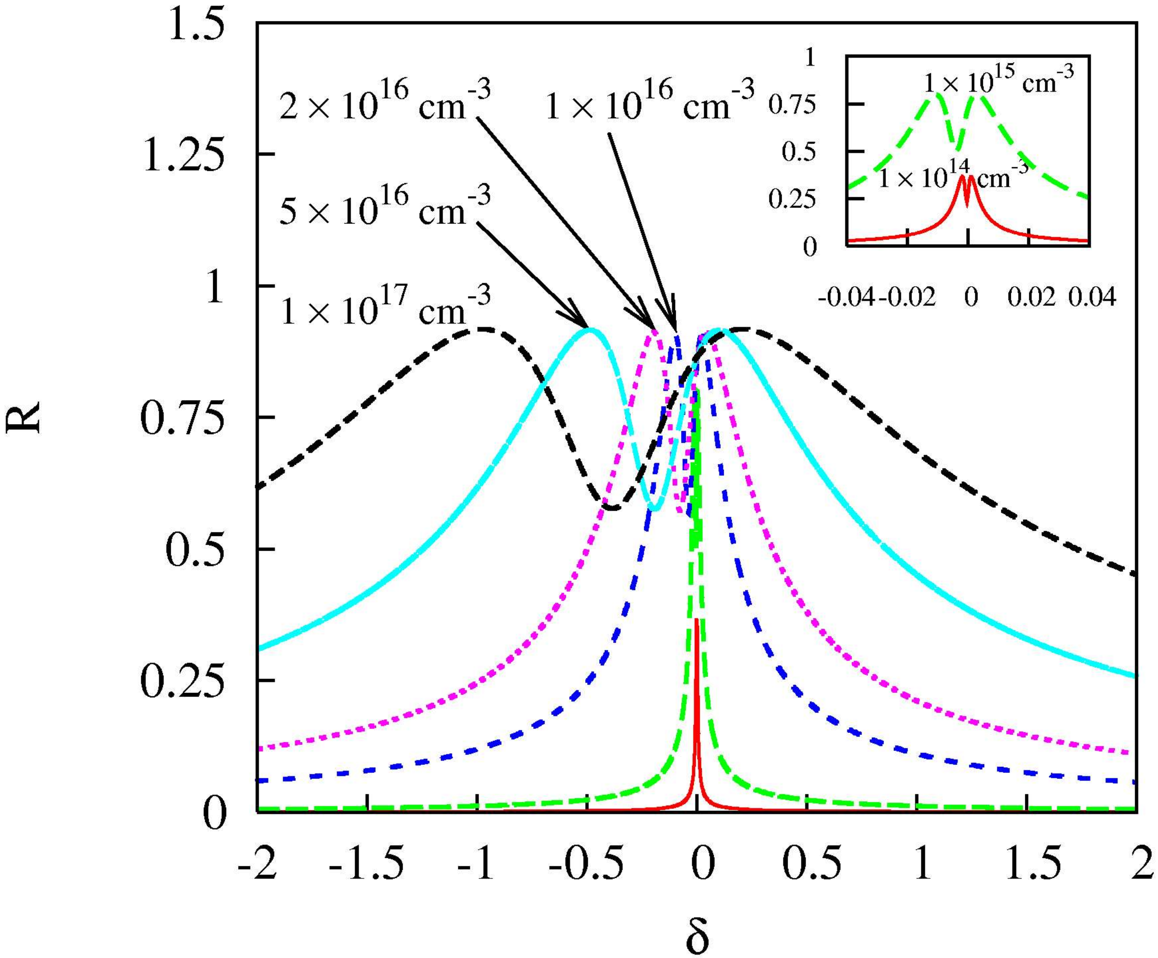
Direct and Cascaded Contributions: Comparison

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

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



$$R = \frac{\left|\chi^{(5)}_{\text{cascaded}}\right|}{\left|\chi^{(5)}_{\text{direct}}\right|}$$

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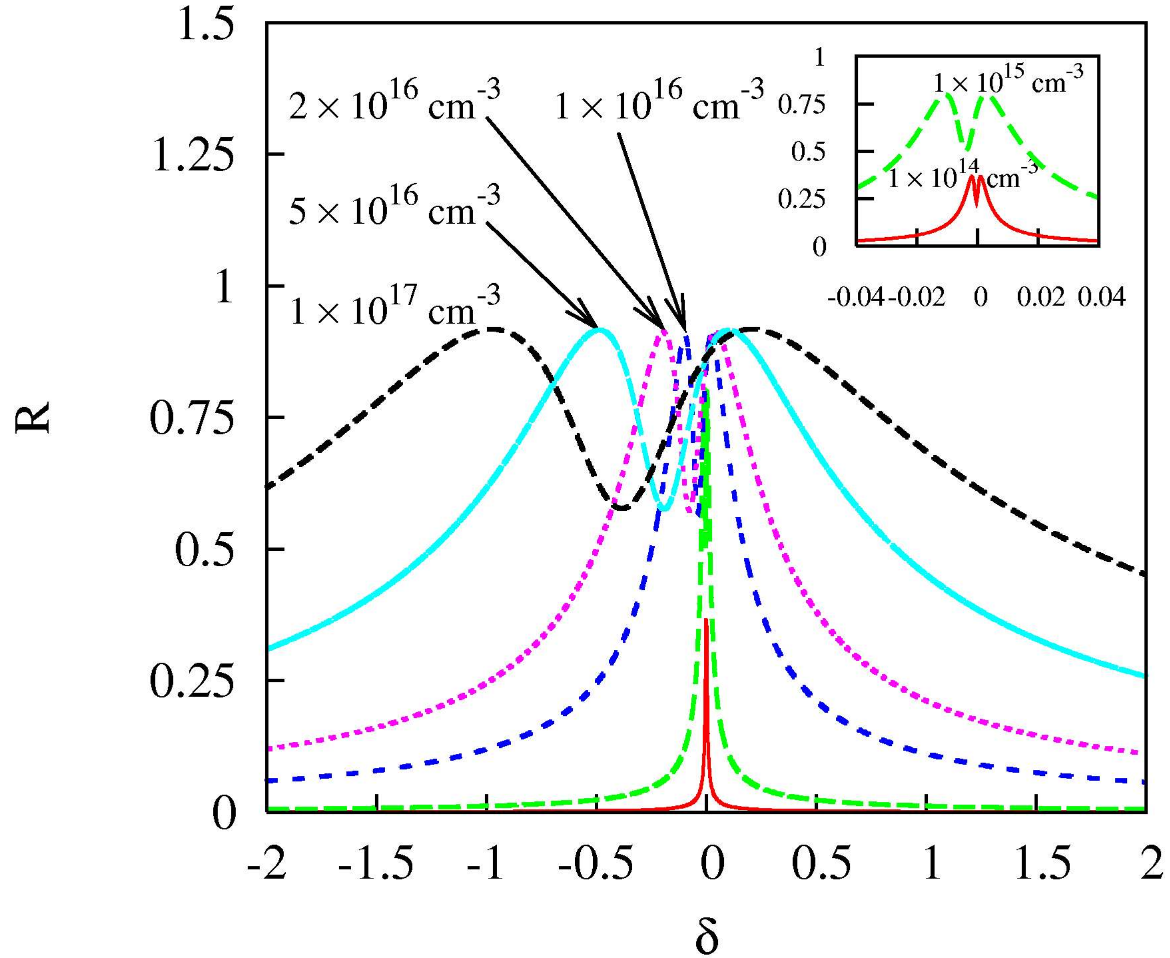
Ratio of absolute values of the contributions as a function of the normalized detuning and atomic density



$$R = \frac{\left|\chi^{(5)}_{\text{cascaded}}\right|}{\left|\chi^{(5)}_{\text{direct}}\right|}$$

Under certain conditions, the cascaded contribution can be as large as the direct contribution.

Conclusions

- Microscopic cascading is possible due to local-field-induced contributions of lower-order nonlinearities to higher-order nonlinearities.
- We demonstrated it based on 5th-order degenerate nonlinearity, following Bloembergen's procedure and Maxwell-Bloch approach.
- 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.

Acknowledgments

- Dr. Sergei Volkov for valuable discussion
- Prof. Boyd's research group