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

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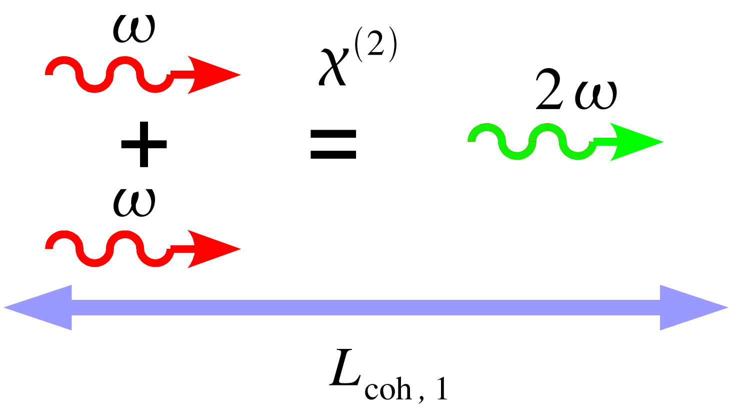
NLP 2007 Wednesday, September 5, 9:30 a. m.

Cascading

In a broad sense:

$$\chi^{(3)}_{\text{eff}} = \text{const} \times \chi^{(2)} : \chi^{(2)}$$

Cascading



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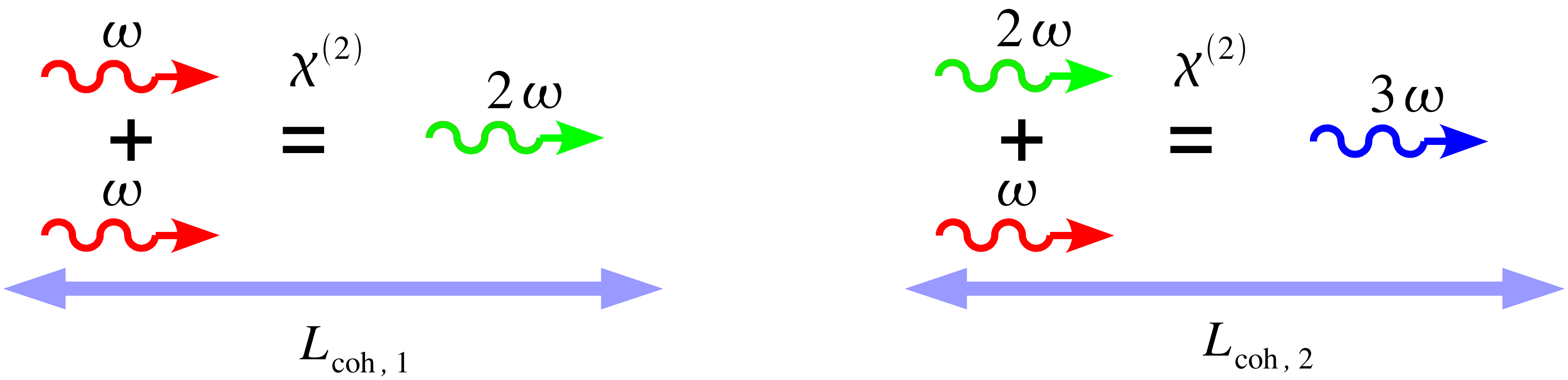
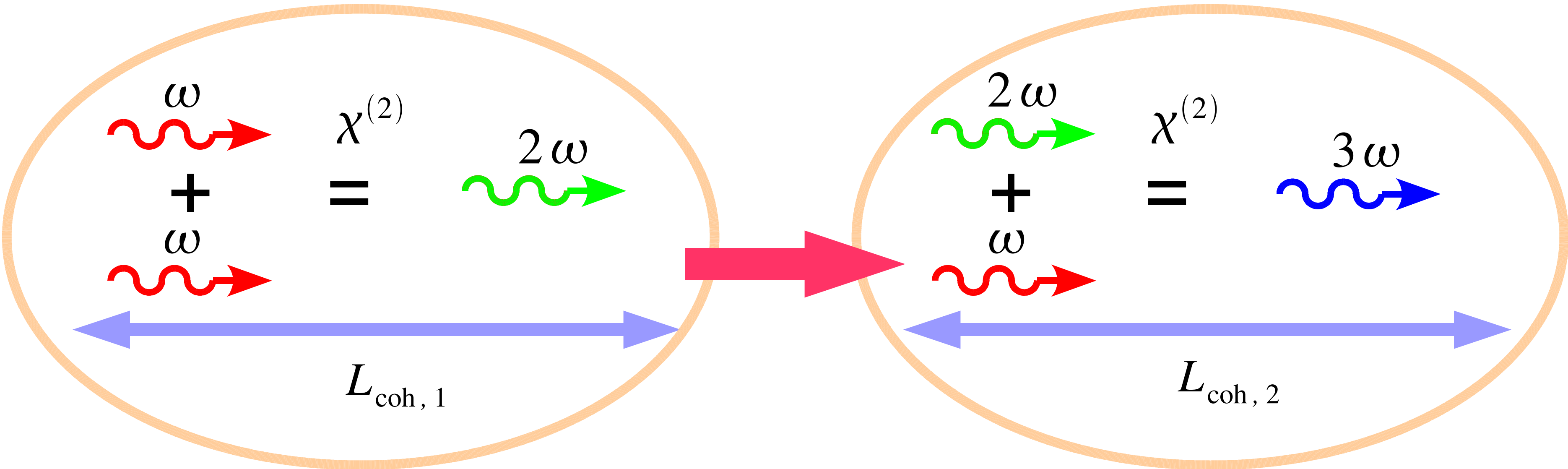


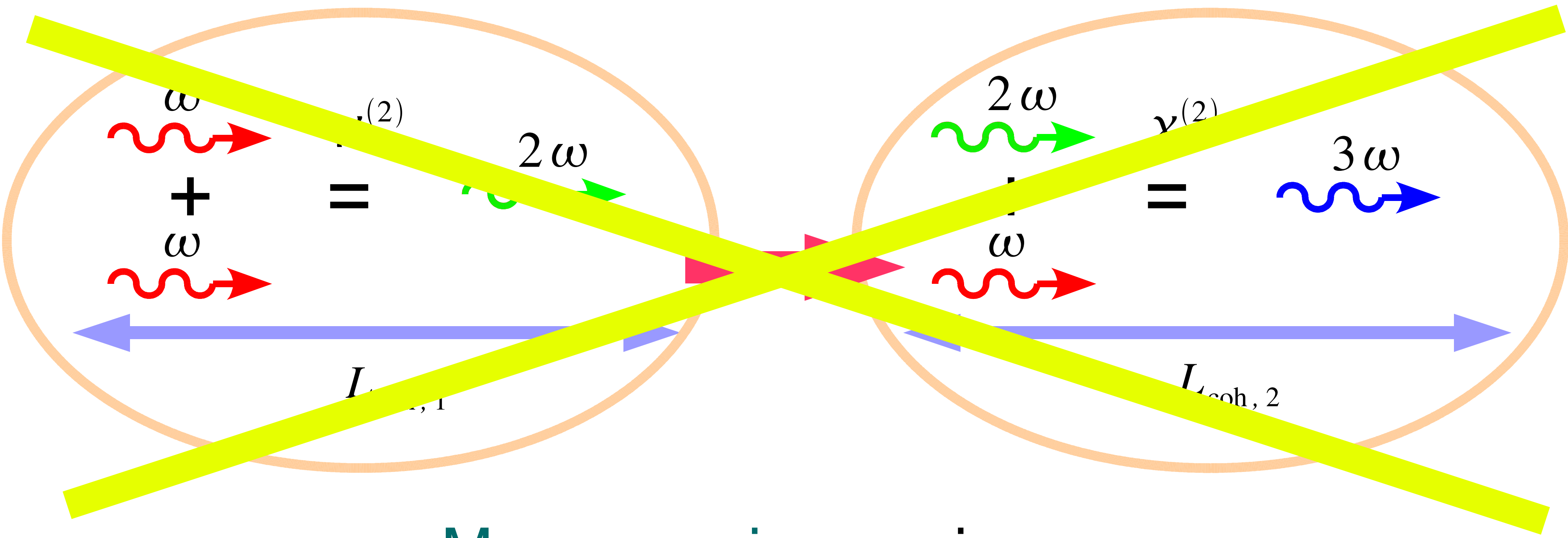
ω
$\chi^{(2)}$
2ω
+
=
ω
$L_{\mathrm{coh},1}$
2ω
$\chi^{(2)}$
3ω
+
=
ω
$L_{\mathrm{coh},2}$

Cascading



Macroscopic: requires propagation and phase-matching

Cascading



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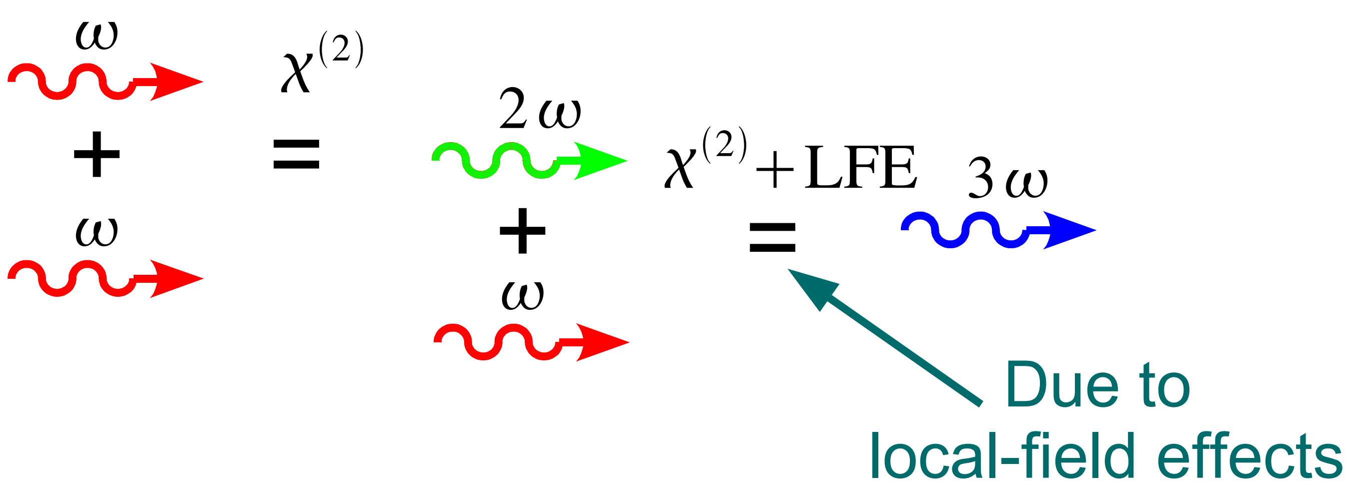

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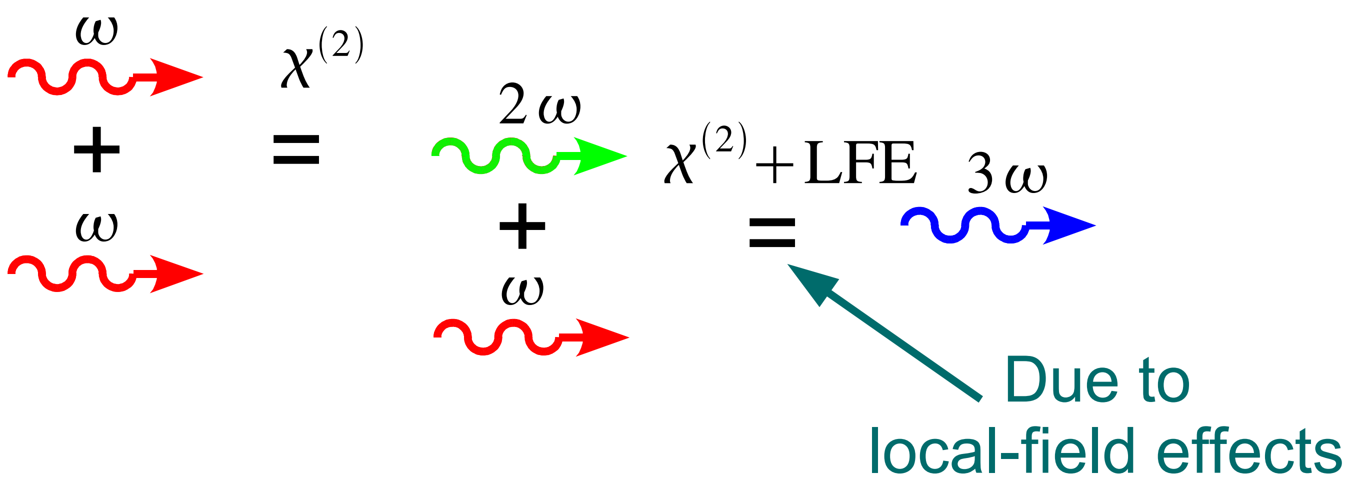
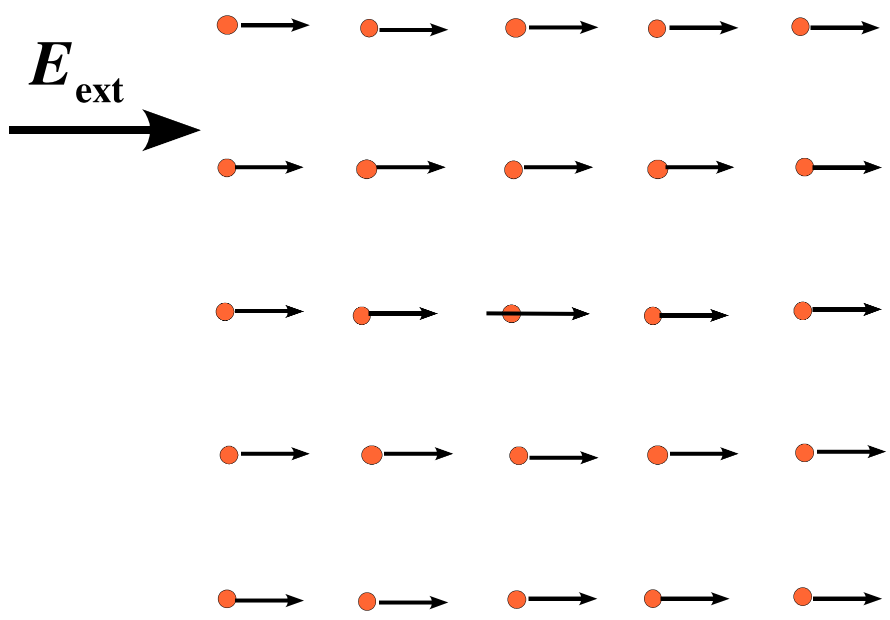


Microscopic: does not require propagation and phase-matching

Lorentz Local Field

Consider a homogeneous medium exposed to an external optical field:



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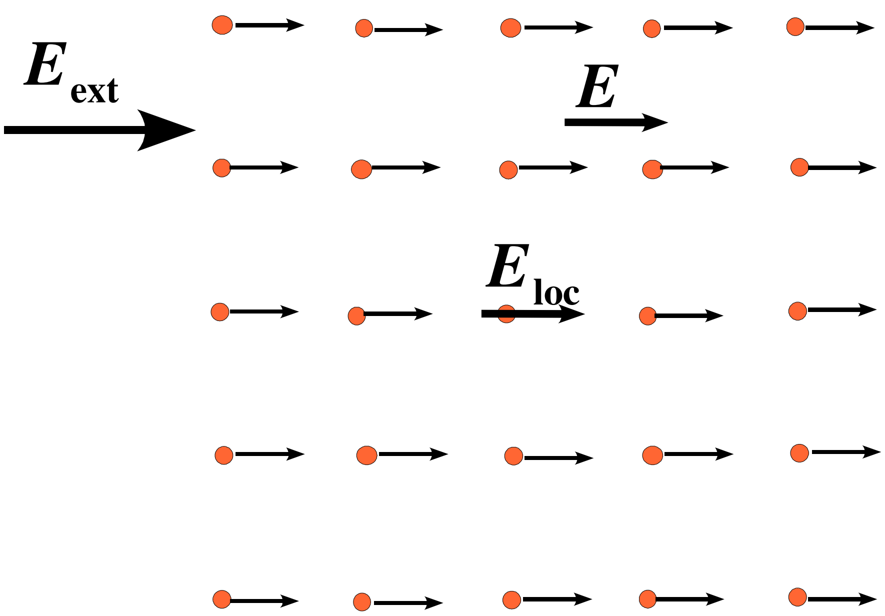
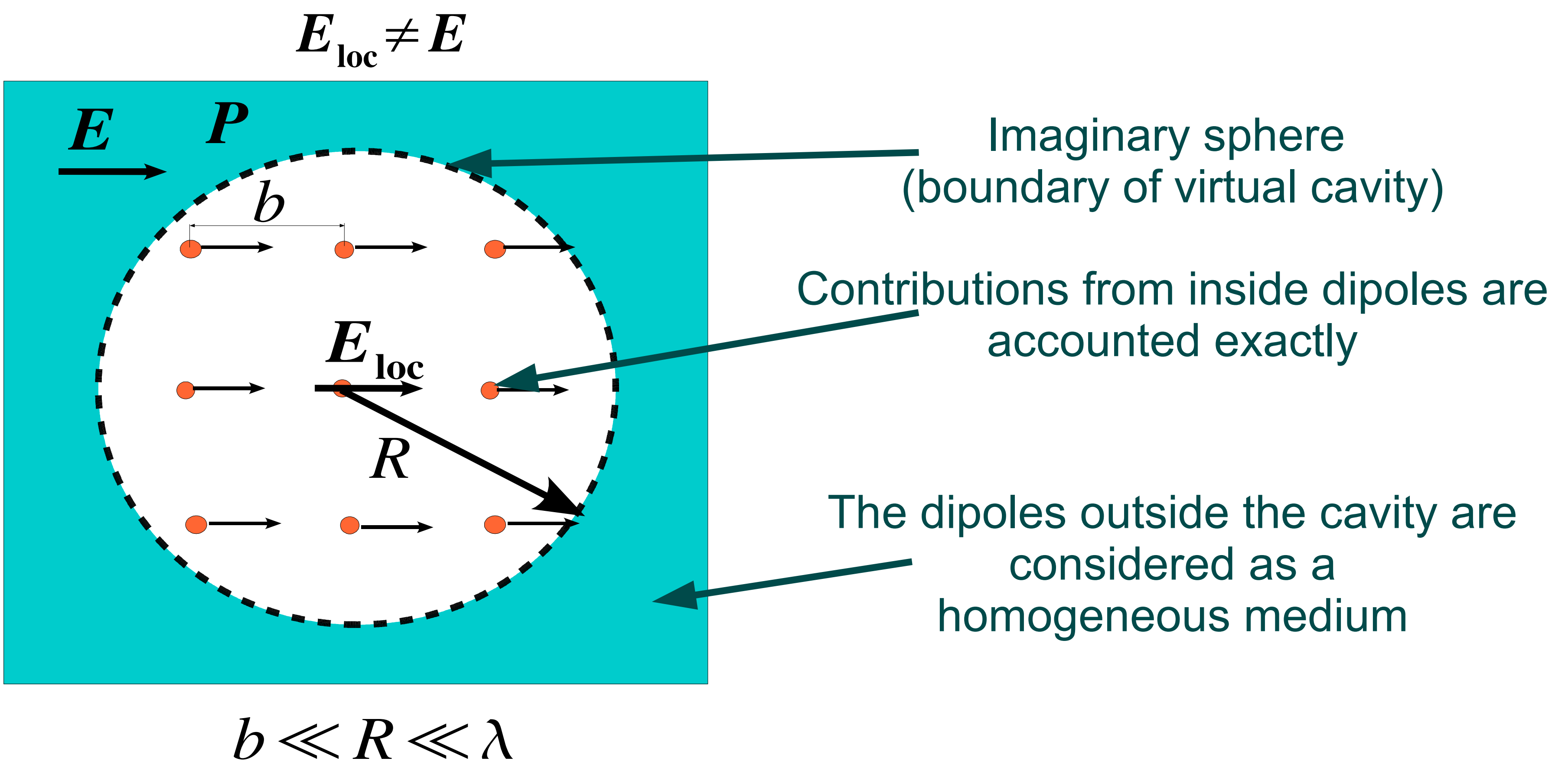


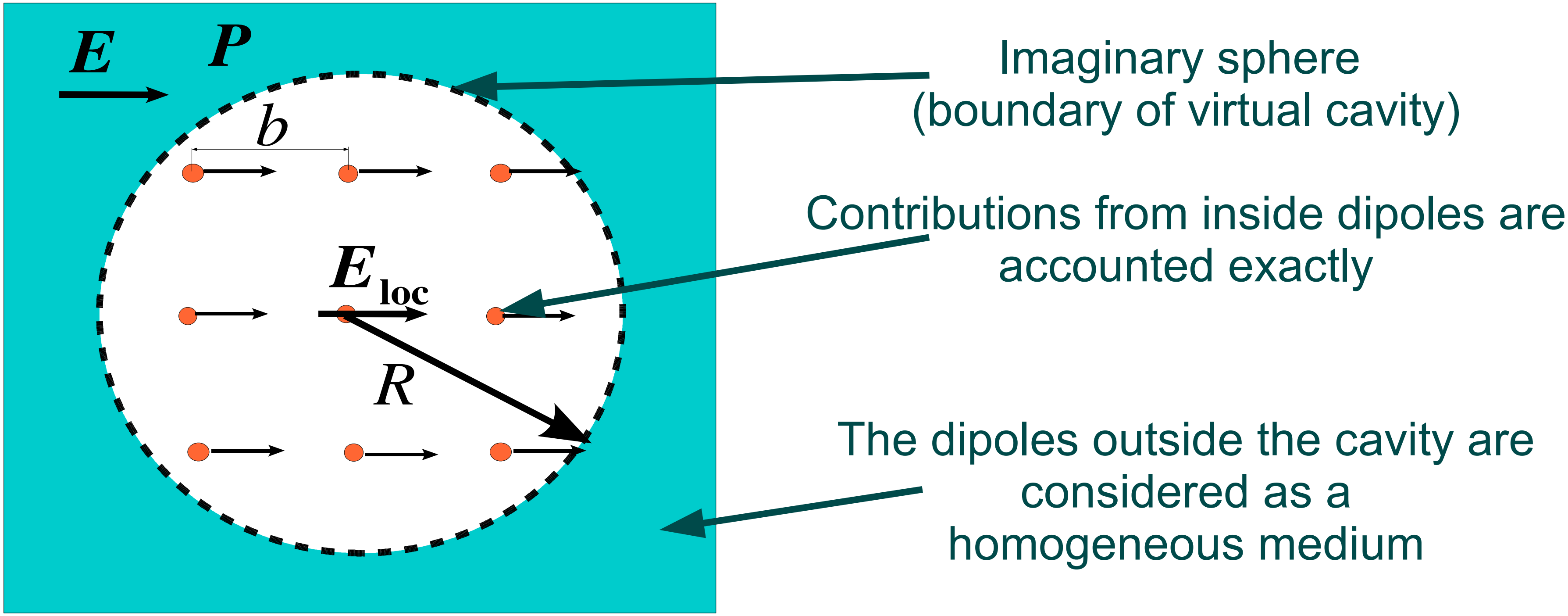
$$E_{\mathrm{loc}} \neq E_{\mathrm{ext}} \neq E$$

Lorentz Local Field



Lorentz Local Field

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



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

$\boldsymbol{E}$ 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

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

Lorentz Local Field

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

or

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

Lorentz Local Field

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

or

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

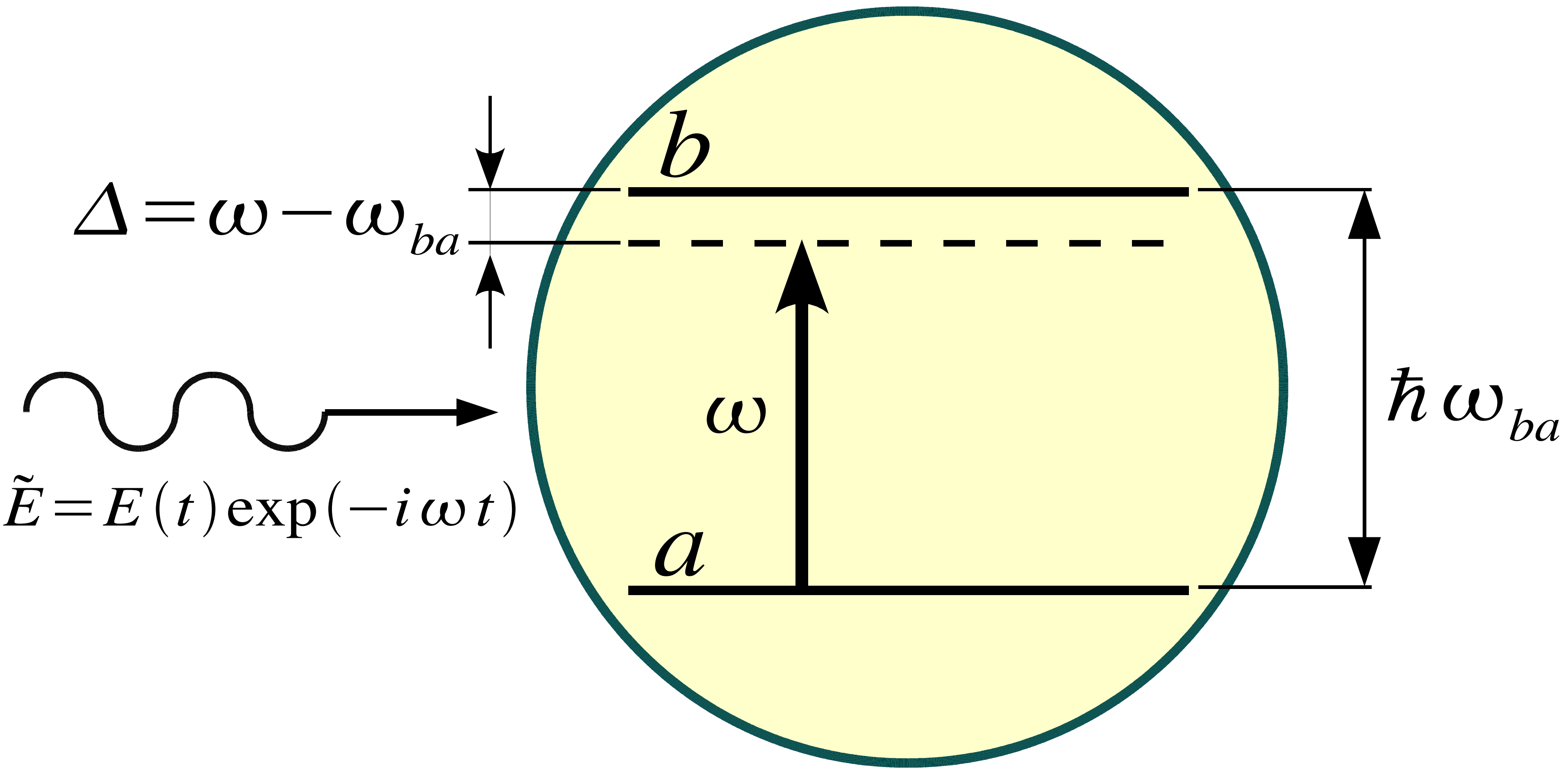
where

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

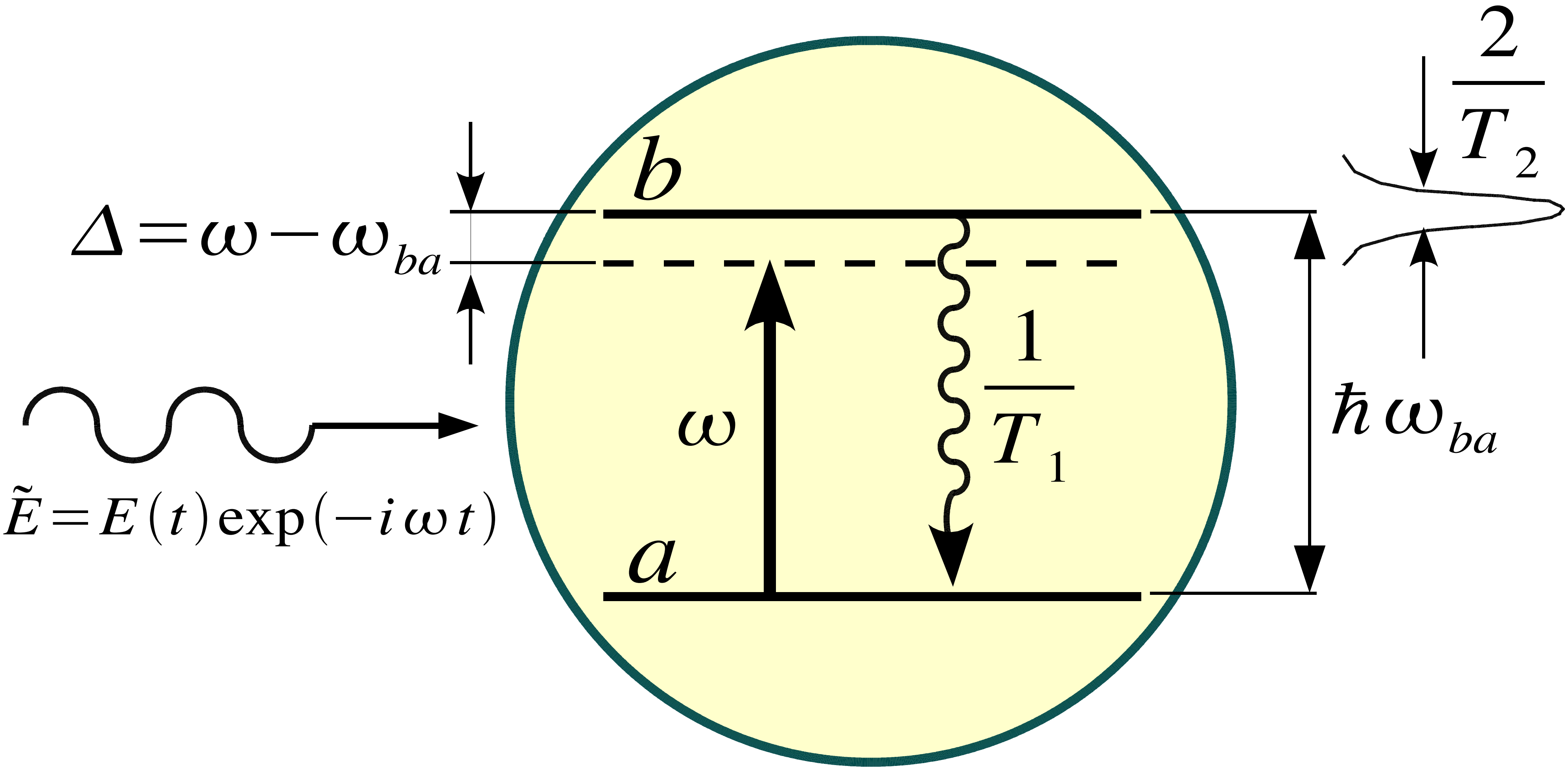
is Lorentz local-field correction factor

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

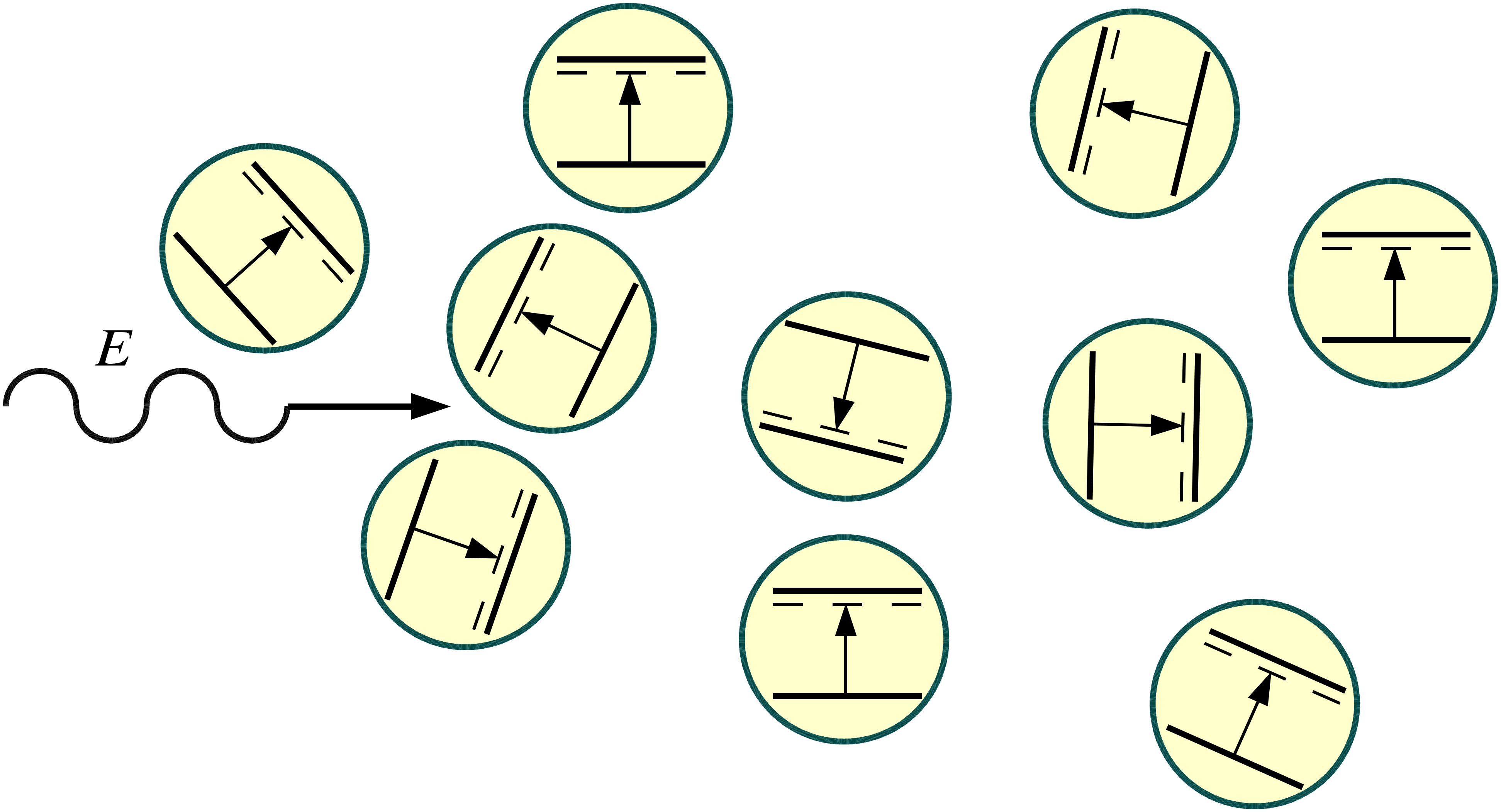
Two-Level Atom



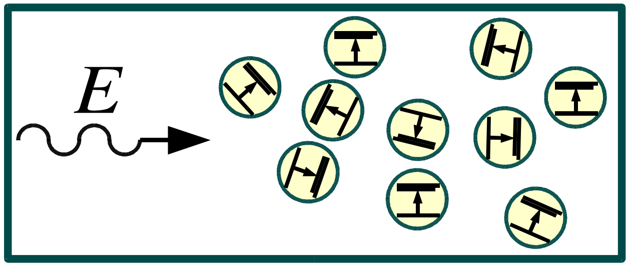
Two-Level Atom



A Collection of Two-Level Atoms



Maxwell-Bloch Equations



$$\dot{\sigma} = \left(i\Delta - \frac{1}{T_2} \right) \sigma - \frac{1}{2} i \kappa E w$$

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

σ is coherence

w is population inversion

w^{eq} is equilibrium population inversion

$\kappa = 2\mu/\hbar$ is atom-field coupling constant

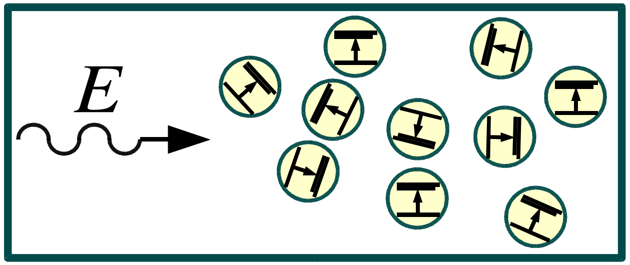
Δ is detuning

μ is transition dipole moment

T_1 is population relaxation time

T_2 is coherence relaxation time

Maxwell-Bloch Equations

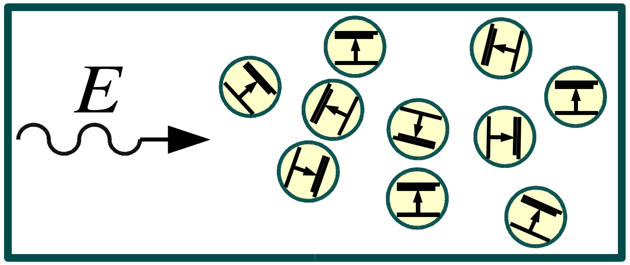


$$E_{\text{loc}} = E + \frac{4\pi}{3} P$$

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

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

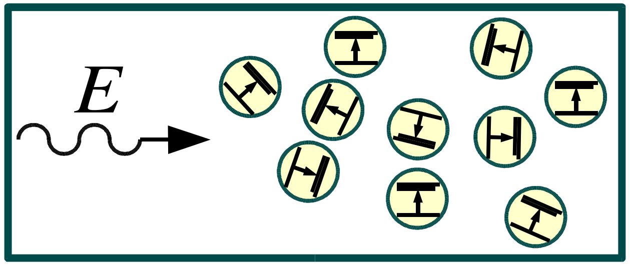
Steady-State Solutions



$$w = -\frac{1}{1 + \dfrac{|E|^2 / |E_s^0|^2}{1 + T_2^2 (\Delta + \Delta_L w)^2}}$$

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

Steady-State Solutions



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inversion-dependent frequency shift

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

Lorentz red shift

Polarization

$$P = N\mu^*\sigma$$

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Local-field-corrected

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Local-field-corrected

Susceptibilities

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

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LF-Corrected Linear and Nonlinear Susceptibilities

The result:

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

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

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

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

$\gamma_{\text{at}}^{(3)}$ and $\gamma_{\text{at}}^{(5)}$ are 3rd- and 5th-order hyperpolarizabilities

LF-Corrected Linear and Nonlinear Susceptibilities

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

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J. D. Jackson, "Classical Electrodynamics"

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

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

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R. W. Boyd, "Nonlinear Optics"

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LF-Corrected Linear and Nonlinear Susceptibilities

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

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

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

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

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

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

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

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

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

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

scales as 6th power of factor L.

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

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 6th power of factor L.

How significant?

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

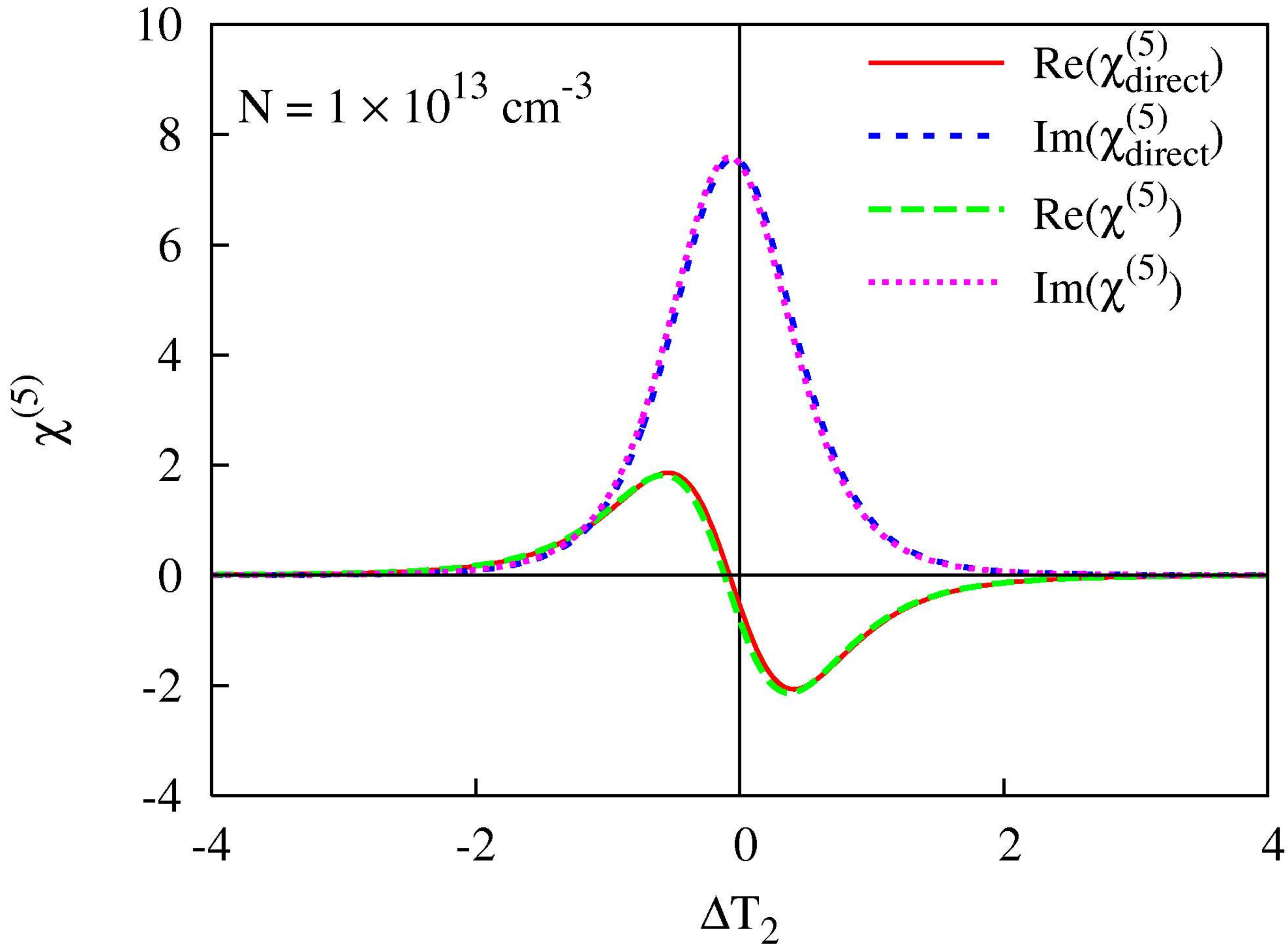
scales as 7th power of factor L.

Example System

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



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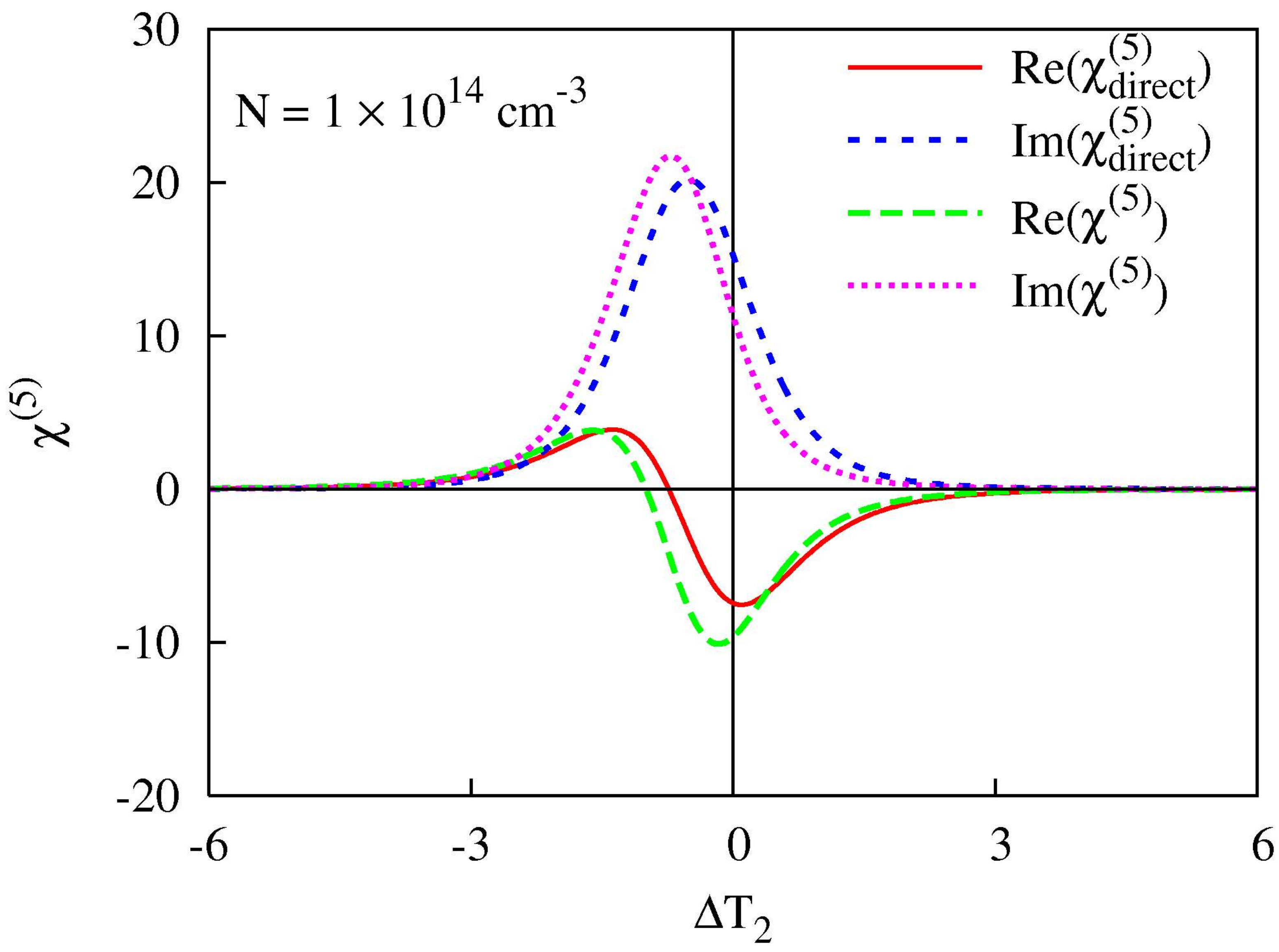

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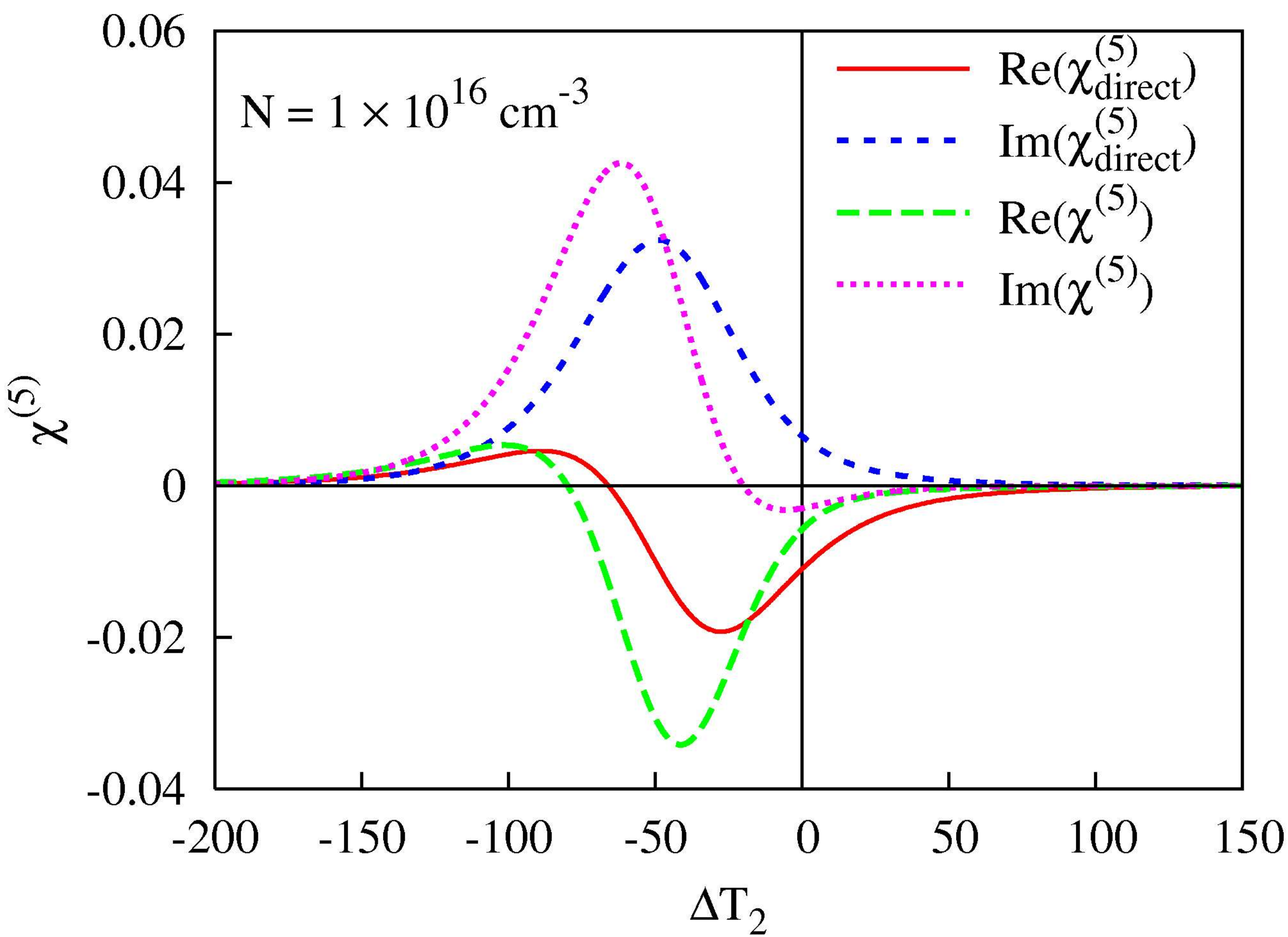

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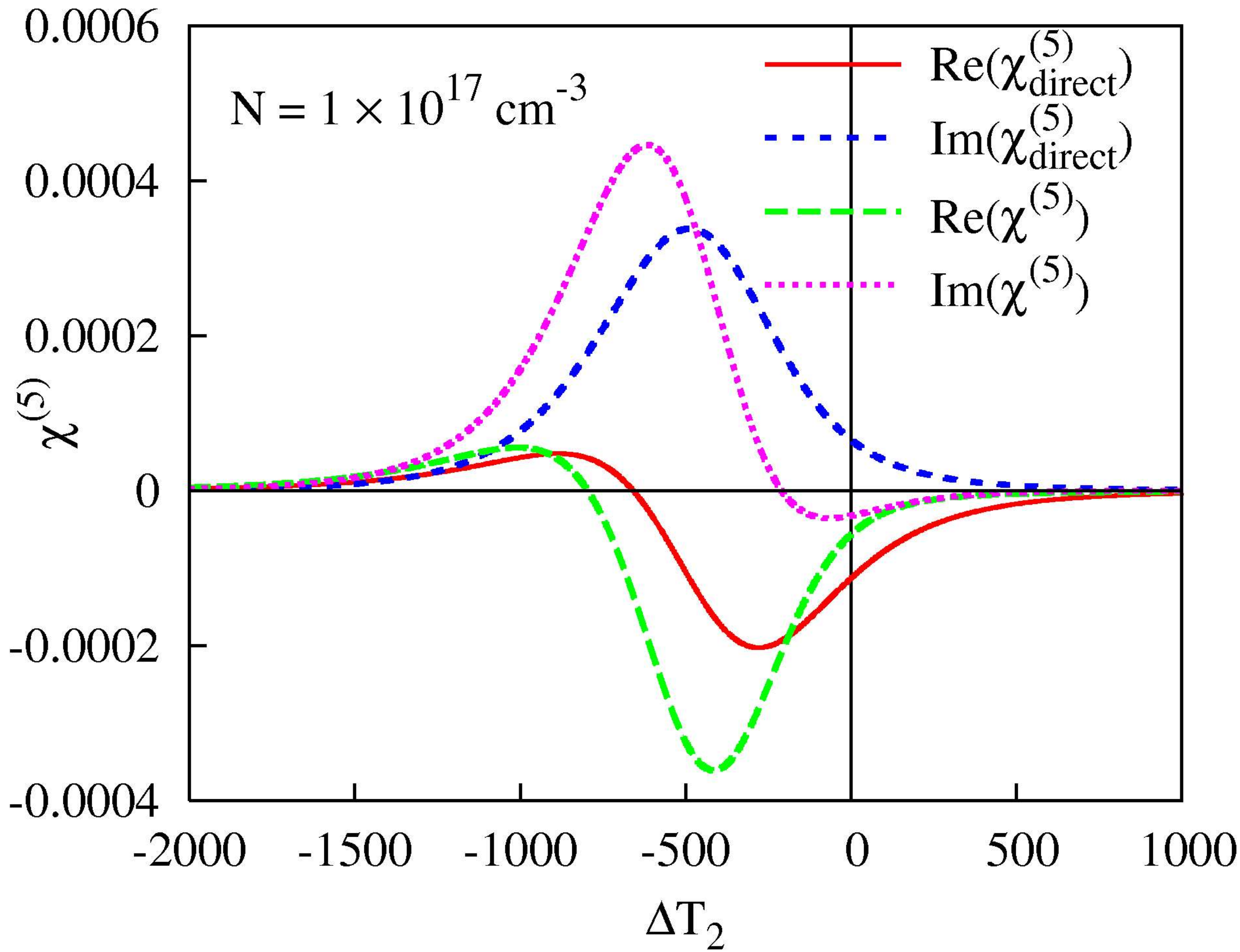

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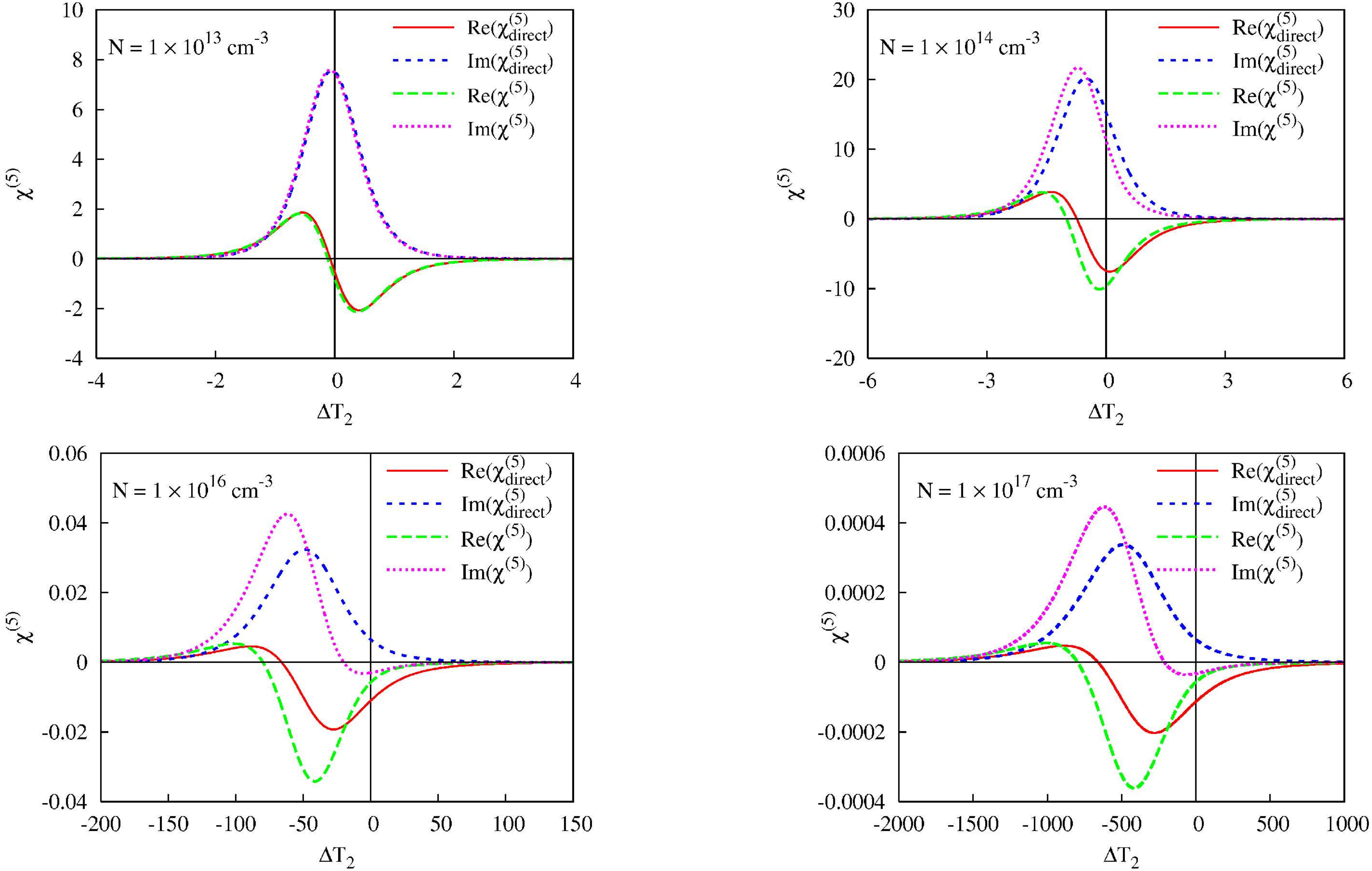
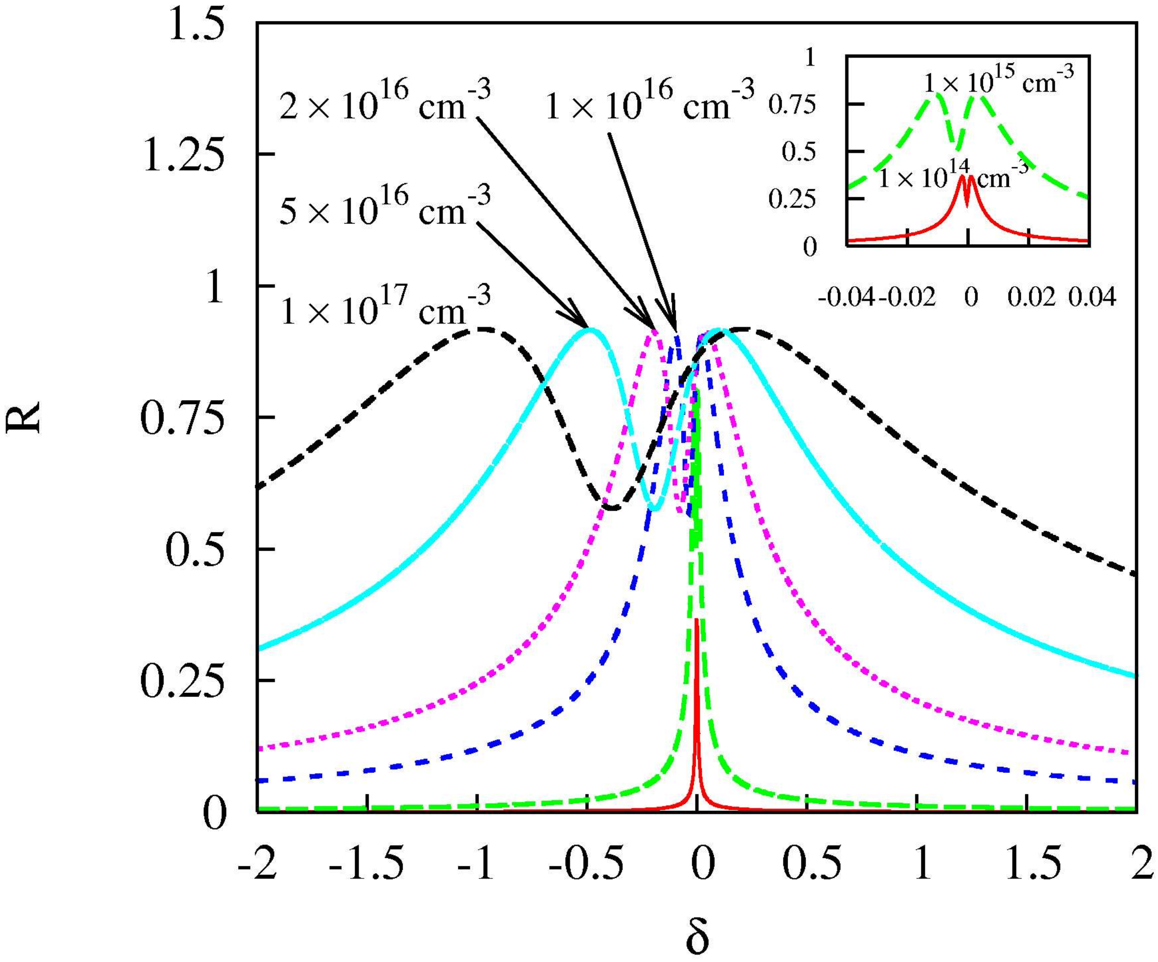


Direct and Cascaded Contributions: Comparison

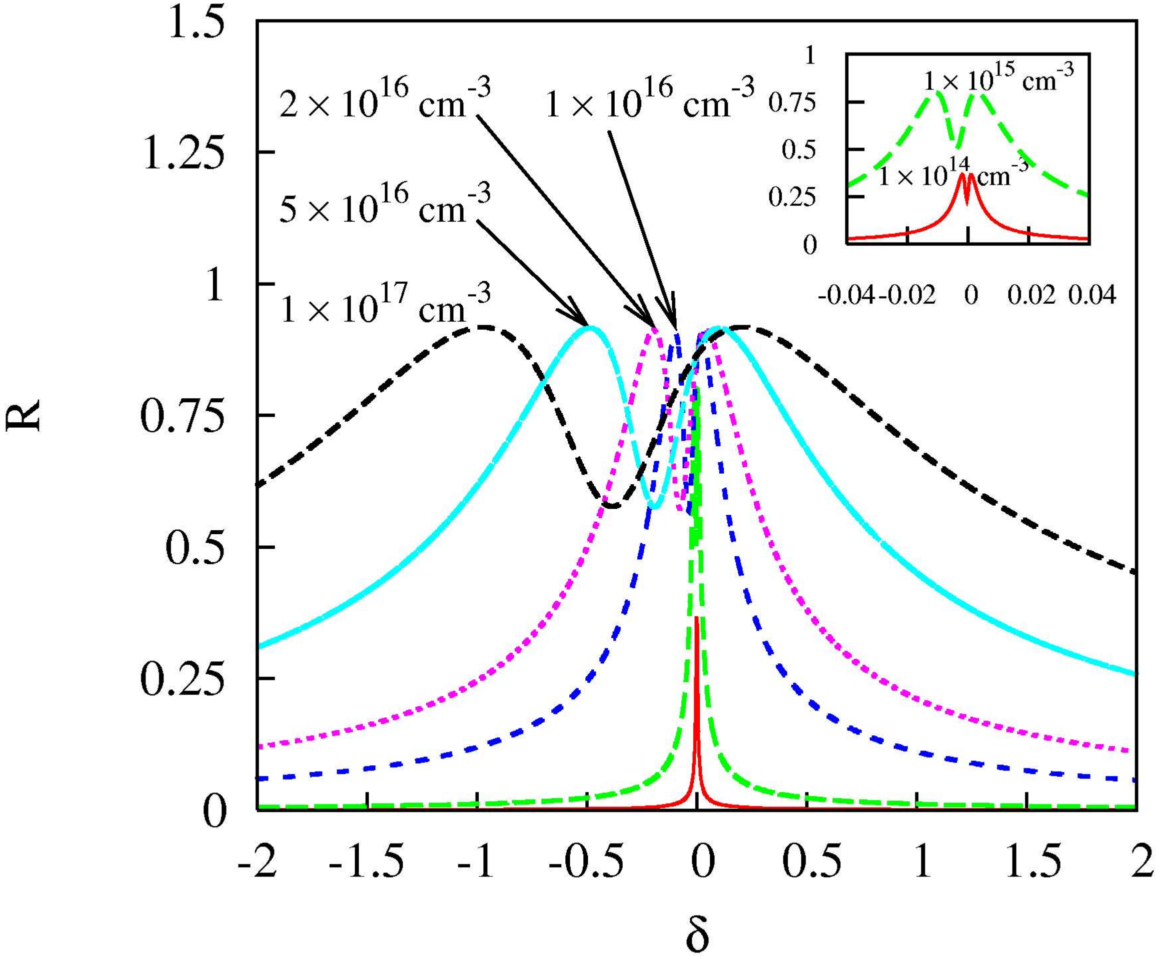
Ratio of absolute values of the contributions as a function of the normalized detuning and atomic density



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

Direct and Cascaded Contributions: Comparison

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



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

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 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.

Acknowledgments

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