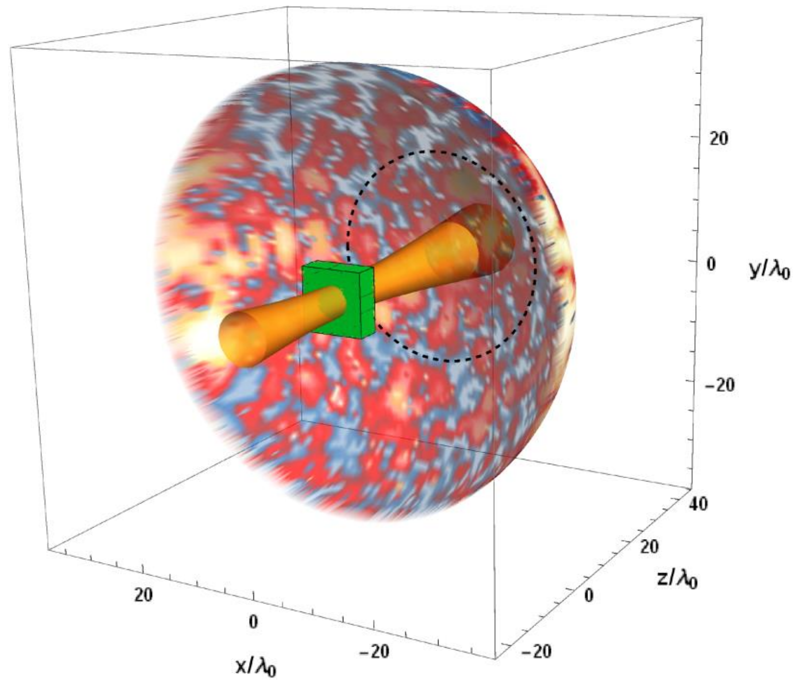

The maximum refractive index of an atomic medium

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Quantum Science Seminar
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frA FUNDACIÓN RAMÓN ARECES

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Univ. Chicago

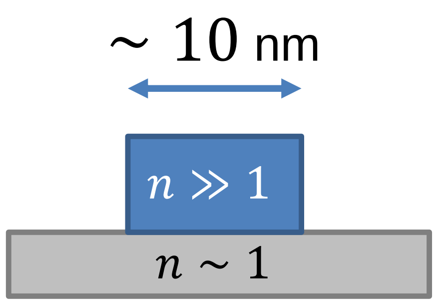
Motivation

- Thousands of optical materials… why is refractive index so always of order unity?

Material	n
Fused silica	1.5
Diamond	2.4
Silicon	3.4
GaAs	3.9
Germanium	4.0

Typical, positive index materials (Telecom/visible wavelengths)

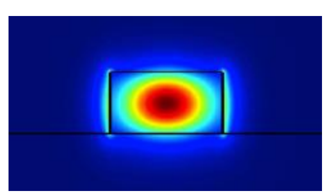
- **Universal physics** governing the value of *n*?
- Profound technological implications if $n \gg 1$ is possible

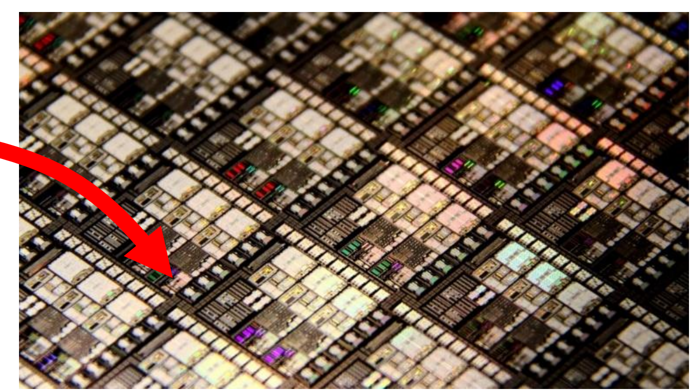


$$\lambda \rightarrow \lambda / n$$

Reduction of scale

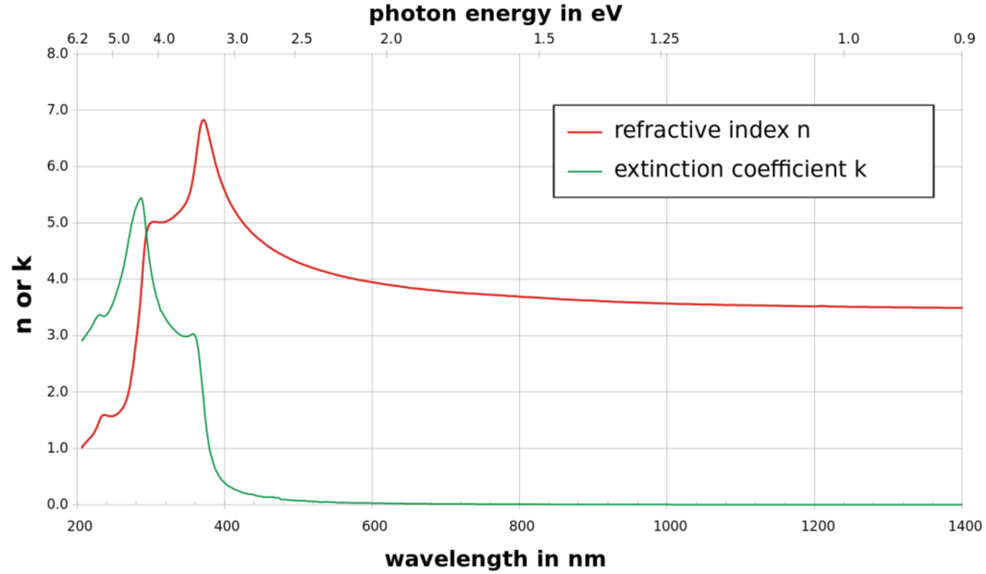
$n \sim 10^2$:





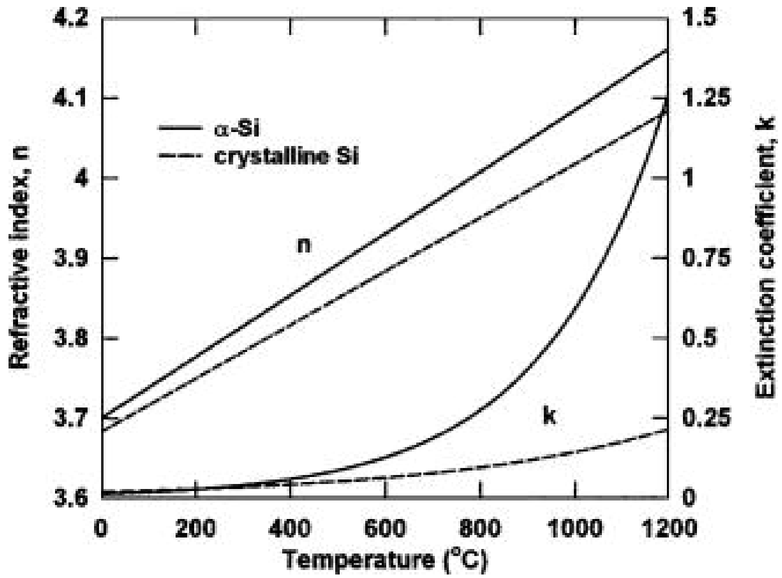
Ruling out possibilities

- We typically work with materials away from their natural electronic resonances?



Index of silicon vs. wavelength

- Phonons?



Index of silicon vs. temperature
J. Appl. Phys. 99, 063516 (2006)

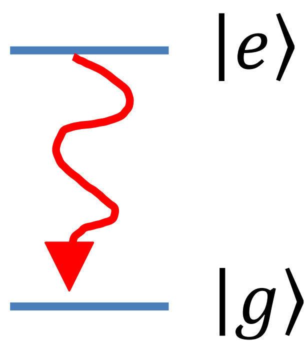
Bottom-up approach

- Minimal model: a “material” near resonance, free of chemistry and solid-state interactions?

Ensemble of well-separated, stationary atoms

- Minimal model of atom:

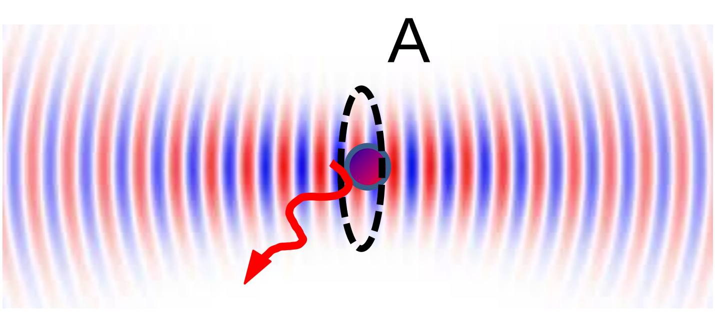
Radiative linewidth Γ_0



Resonant wavelength λ_{eg}

- Optical response of a single atom:

$\boxed{\lambda_{eg} \sim 1\ \mu\text{m}, \quad \text{Bohr radius} \sim 0.1\ \text{nm}}$

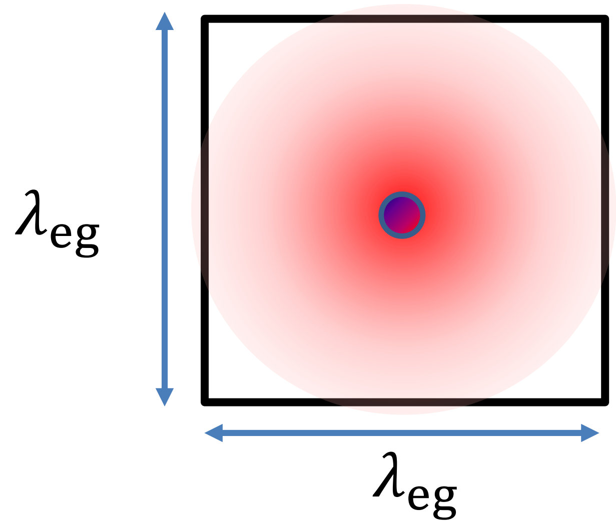


$$\text{Scattering probability} \sim \frac{\lambda_{\text{eg}}^2}{A}$$

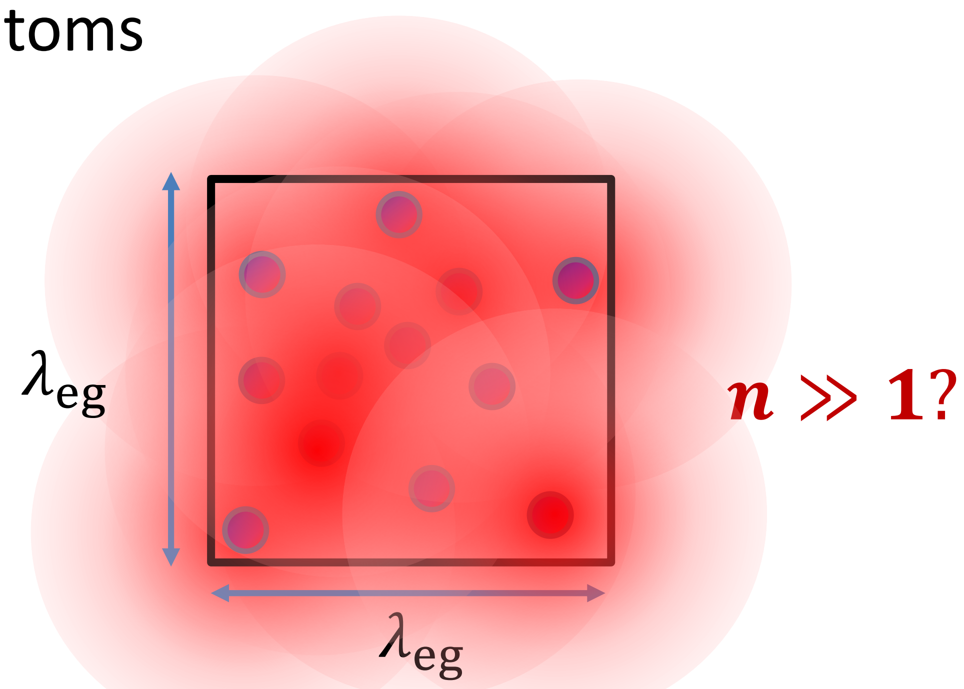
Extraordinary response compared to physical size!

Index of an atomic medium

- Single atom



- Ensemble of atoms

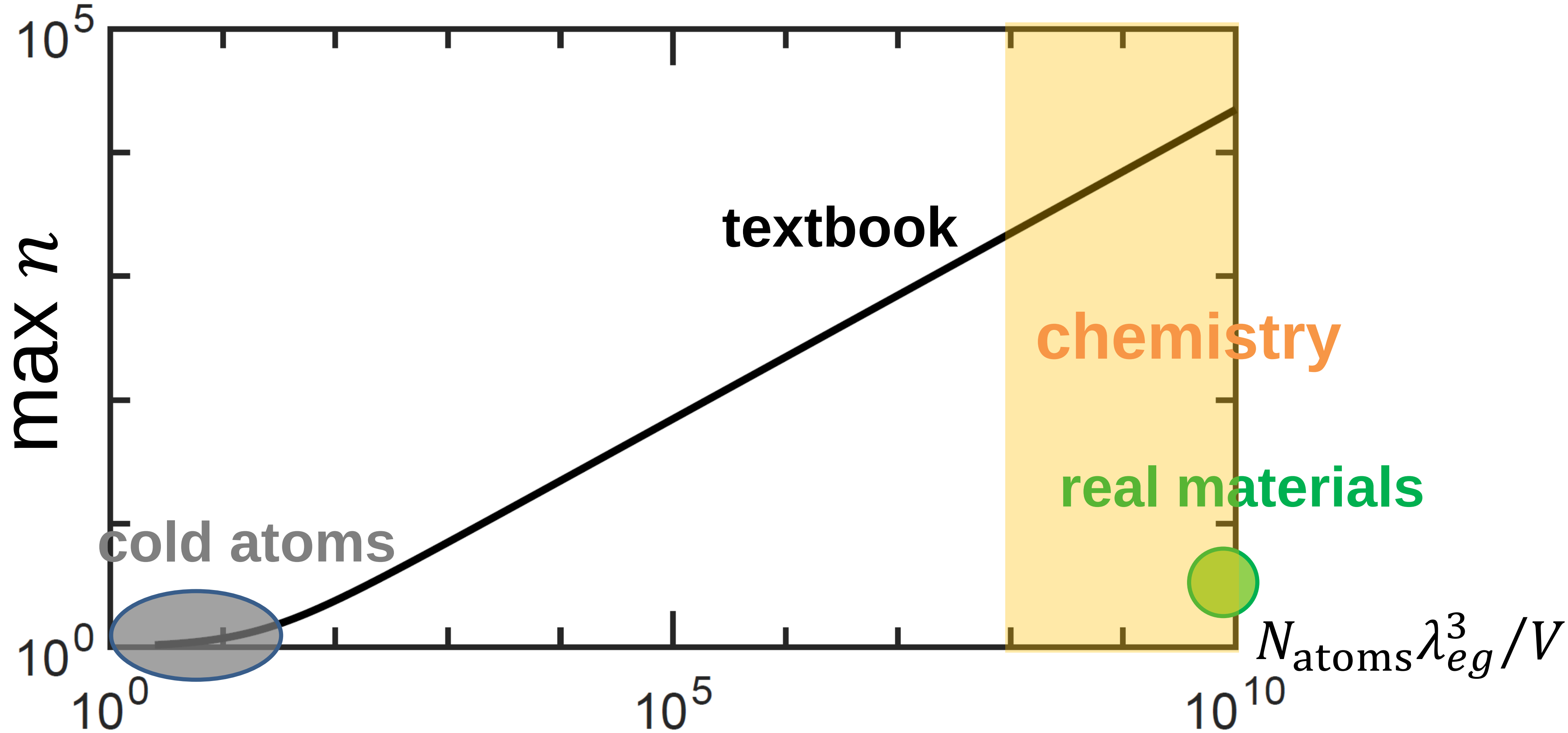


Index of an atomic ensemble

- Drude-Lorentz / Maxwell-Bloch equations:

$$n_{\mathrm{MB}}(\omega)^2 = \epsilon_{MB}(\omega) \sim 1 + \alpha(\omega)\frac{N_{\mathrm{atoms}}}{V}$$

- Problem!

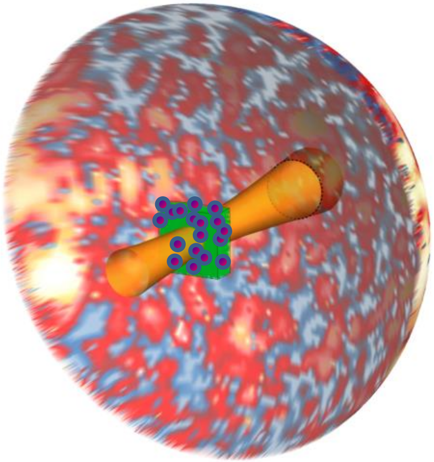


What's missing in the textbook answer?

$$n_{\mathrm{MB}}(\omega)^2 \sim 1 + \alpha(\omega)\frac{N_{\mathrm{atoms}}}{V}$$

- Hints:
  - Doesn't depend on microscopic configuration
  - Multiplicative (every atom does the same thing)
- **Missing:** wave interference and multiple scattering
- Light propagation through a random medium is complex!

Outgoing "speckle" intensity pattern

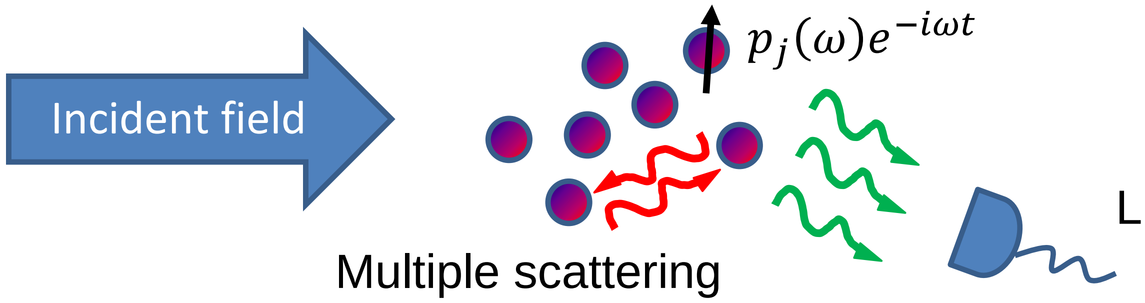


- These features tend to get suppressed for dilute, moving atoms

Another approach: coupled dipoles

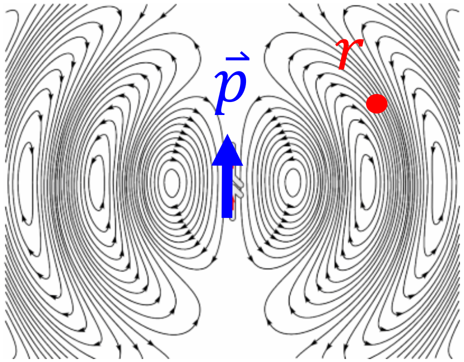
- Collection of (classical) polarizable dipoles:

$E_{in}(r,\omega)e^{-i\omega t}$



- Total field: $$E(r,\omega) = E_{in}(r,\omega) + \sum_j G(r,r_j,\omega)\,p_j(\omega)$$

- In free space:



$$G(r,0,\omega) = e^{ikr}\left[\frac{k^2}{r}(\hat{n}\times\hat{p})\times\hat{n} + (3\hat{n}(\hat{n}\cdot\hat{p}) - \hat{p})\left(\frac{1}{r^3} - \frac{ik}{r^2}\right)\right]$$

$$k = \omega/c$$

- Solve for dipoles: $p_i = \alpha(\omega)E(r_i,\omega) = E_{\text{in}}(r_i,\omega) + \sum_j G(r_i,r_j,\omega)\,p_j$

- Quantum version?

Optical properties of dense atomic gases

- Significant body of work already on dense atomic gases

Theory J. Ruostekoski, S. Yelin, M. Fleischhauer, J.-J. Greffet, A.M. Rey, S.E. Skipetrov, T. Wellens, O. Romero-Isart…

Experiment A. Browaeys, R. Kaiser, C.S. Adams, J. Ye, J. Beugnon, …

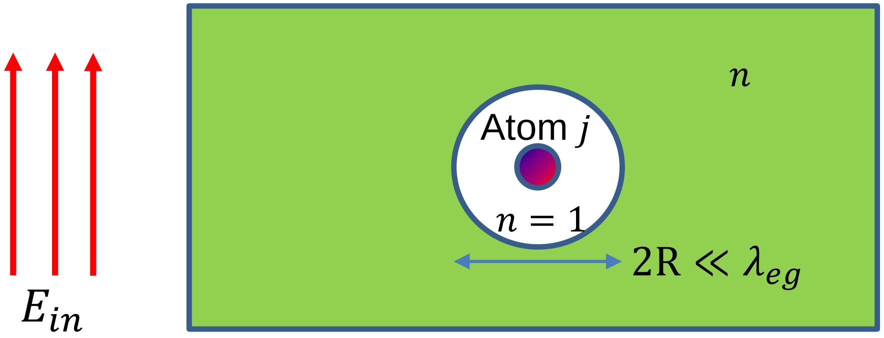
- General agreement that one must go beyond Maxwell-Bloch equations
  - Including observations of smaller optical response
- But a clear-cut answer is still elusive!

Challenges of coupled dipole approach:

- Number of equations $\propto N_{\text{atoms}}$
- Too much information!

Lorentz-Lorenz model

- Famous approximate model for multiple scattering: Lorentz-Lorenz
- Atom j sits in a small shell of vacuum, surrounded by a smooth index provided by other atoms



$$p_j \approx \alpha(\omega)\left[E_{in}(r_j, \omega) + \frac{n(\omega)^2 - 1}{3} E_{in}(r_j, \omega)\right]$$

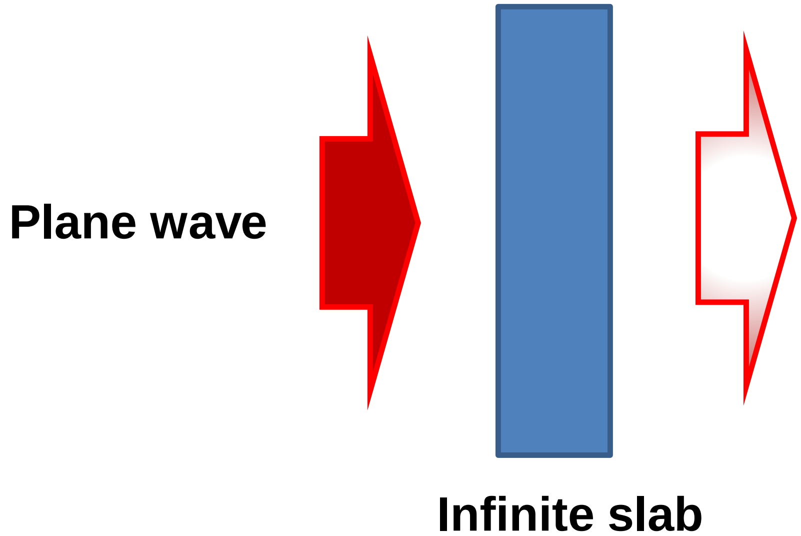
- Lorentz-Lorenz still seems insufficient for near-resonant atoms

$$n_{LL}(\omega) = n_{MB}(\omega + N_{\text{atoms}}\lambda^3/8\pi^2 V)$$

Questions?

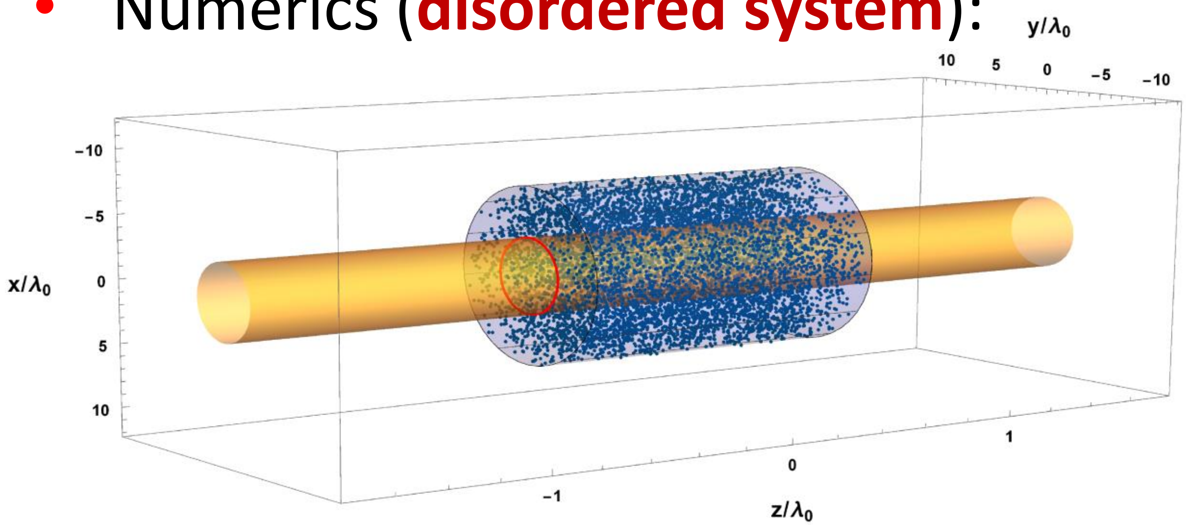
Numerical approach

- Conceptual idea:



$$E_{out} = t(\omega)E_{in} \sim e^{i(\omega/c)nL}E_{in}$$

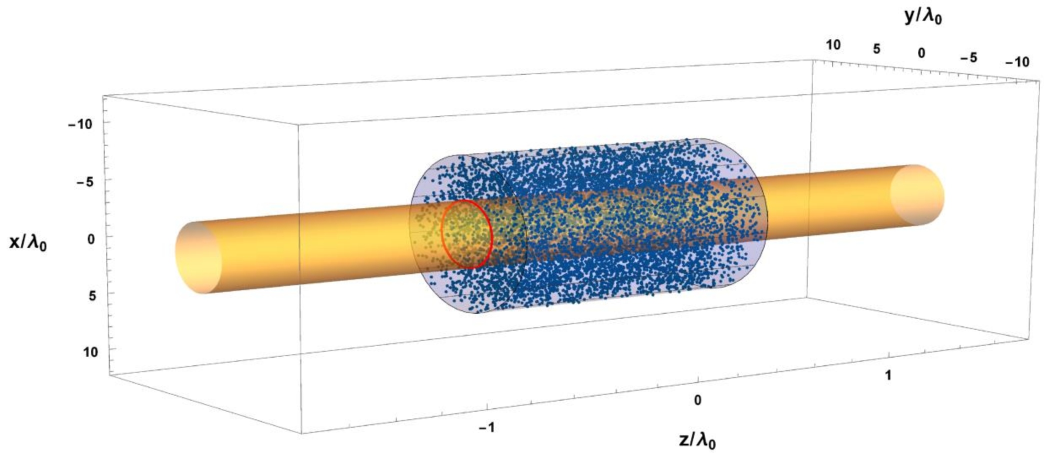
- Numerics (**disordered system**):



Up to $\mathrm{N}_{\mathrm{atoms}} \sim 3 \times 10^4$ atoms randomly distributed
Averaging over configurations

- Focused Gaussian input ($w_0 < R_{\mathrm{cyl}}$) to avoid diffraction effects

Numerical approach



- Solve coupled dipole equations

$$\langle \mathbf{Gaussian}| \quad E(r,\omega) = E_{in}(r,\omega) + \sum_j G(r, r_j, \omega)\, p_j(\omega)$$

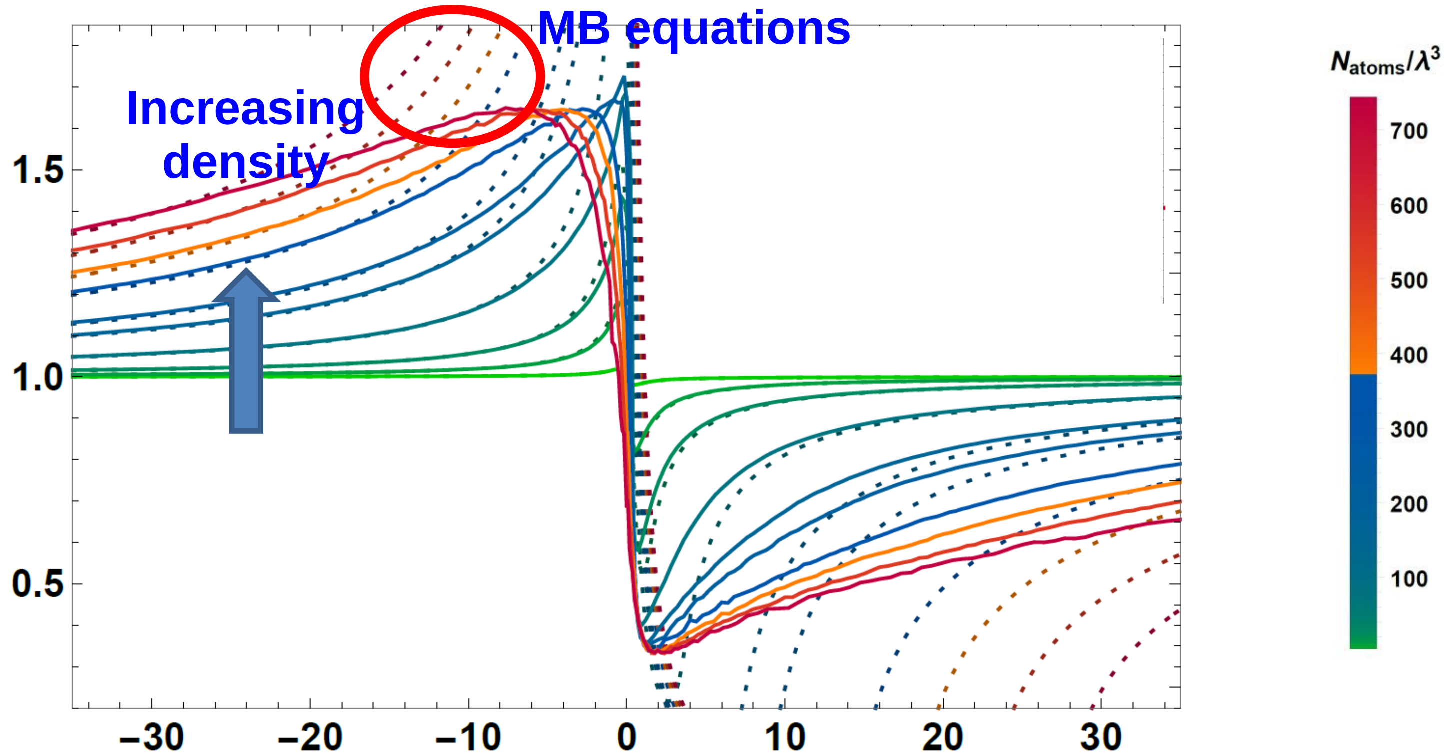
- Point-by-point field re-construction:
  - Time-consuming, and contains complex "speckle" pattern

- Project transmitted field back into the Gaussian mode

$$t \sim 1 + i\left(\frac{\lambda_0^2}{w_0^2}\right)\sum_j \frac{E_{in}^*(r_j)}{E_{\text{in,max}}} p_j \sim e^{ik_0 n L}$$

Numerical results

Re(n) vs. frequency

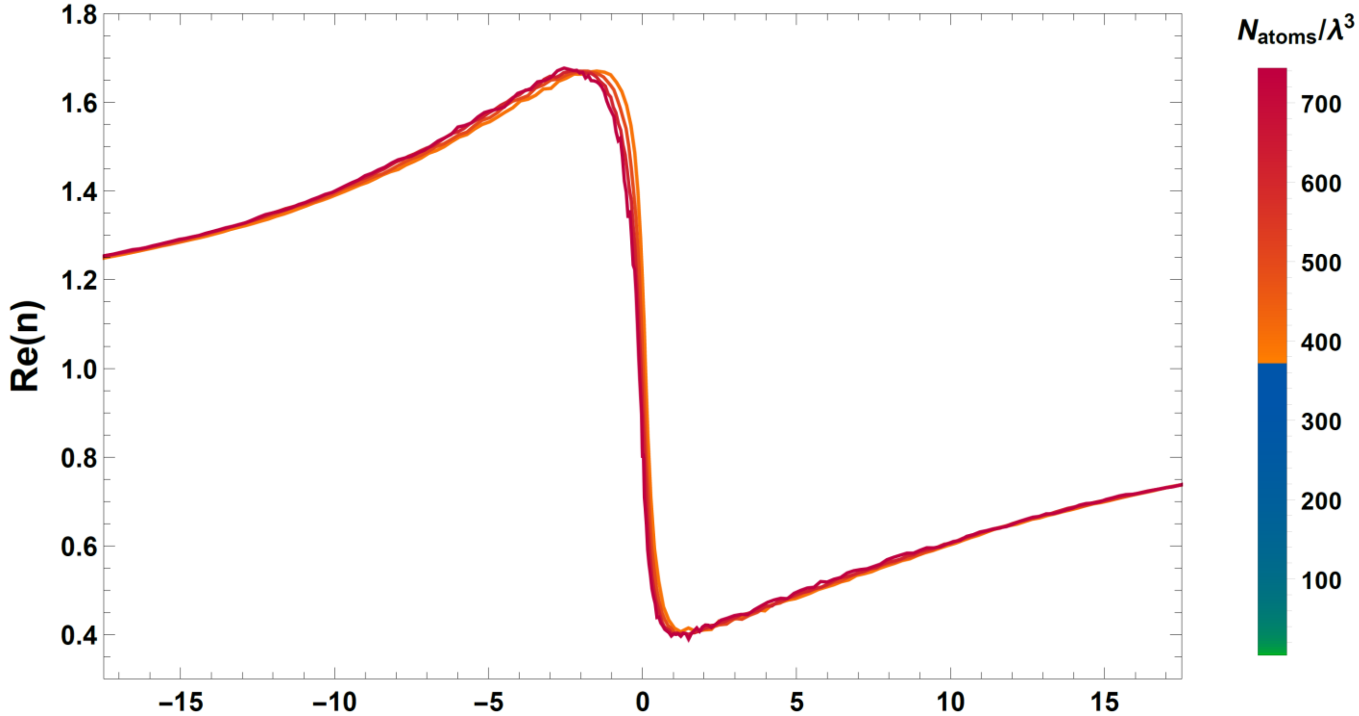


$\Delta/\Gamma_0 \quad (\Delta = \omega_{in} - \omega_{eg})$

- Clear deviation from MB equations for $N_{\text{atoms}}/\lambda^3 > 10^2$

Scale invariance

Re(n) vs. rescaled frequency



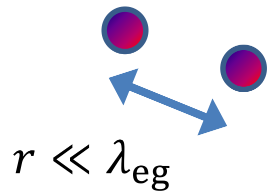
$\Delta\lambda^3/\Gamma_0 N_{\text{atoms}}$

Key points:

- Invariant rescaled spectrum $n\left(\omega\lambda^3/\Gamma_0 N_{\text{atoms}}\right)$ at high densities
- Maximum index saturates to a "real-life" value of $n \approx 1.7$!!
- Saturation occurs at densities $\sim 10^8$ times less dilute than a solid

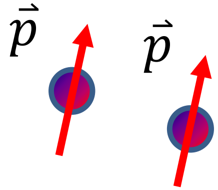
A simple theory: two atoms

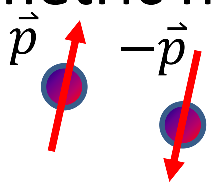
- Dipoles strongly interact via their near fields



$G(r_1, r_2) \sim 1/r^3$

- Symmetric and anti-symmetric normal modes $|\pm\rangle$



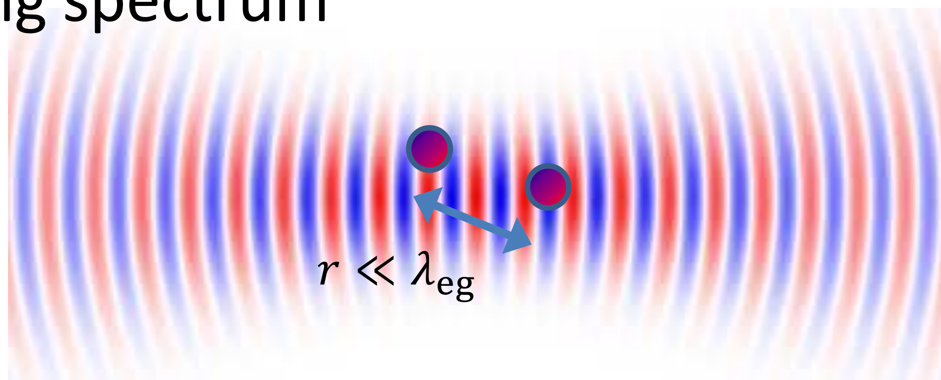


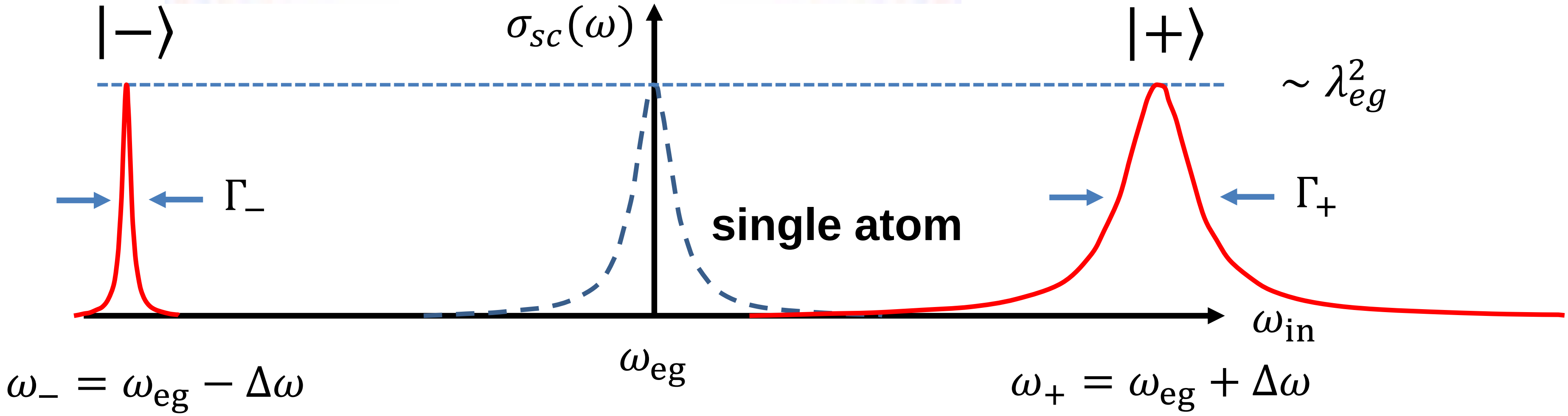
$\Gamma_+ \approx 2\Gamma_0$ $\quad$ $\Gamma_- \approx \Gamma_0 \left(r/\lambda_{eg}\right)^2$ **Dicke sub/superradiance**

$\Delta\omega_+ \sim \pm\Gamma_0 \left(\lambda_{eg}/r\right)^3$ $\quad$ $\Delta\omega_- \sim \mp\Gamma_0 \left(\lambda_{eg}/r\right)^3$ **Strong frequency shift**

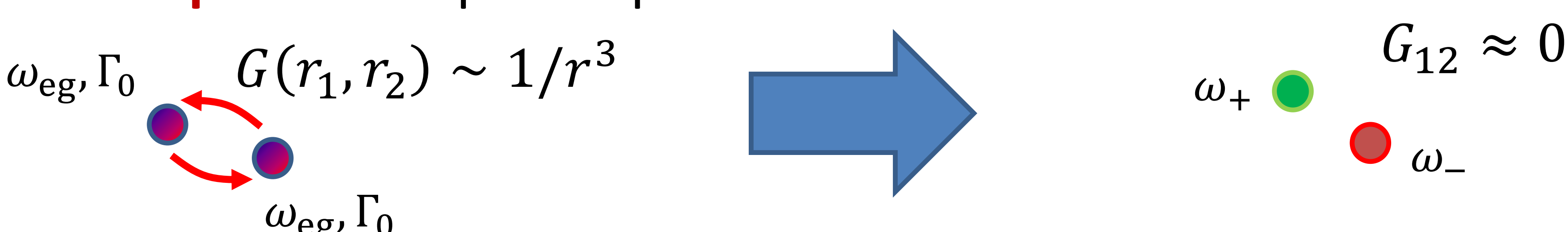
A simple theory: two atoms

- Scattering spectrum





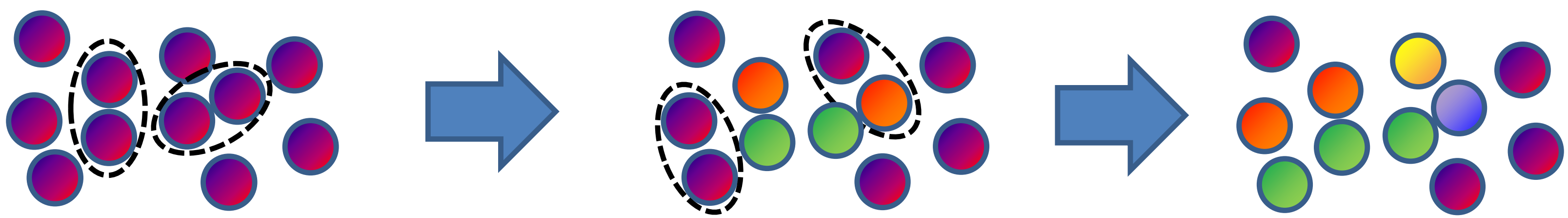
- **Equivalent** optical spectrum



Two inhomogeneous, non-interacting atoms!

Many atoms: renormalization group

- Disordered atomic ensemble:

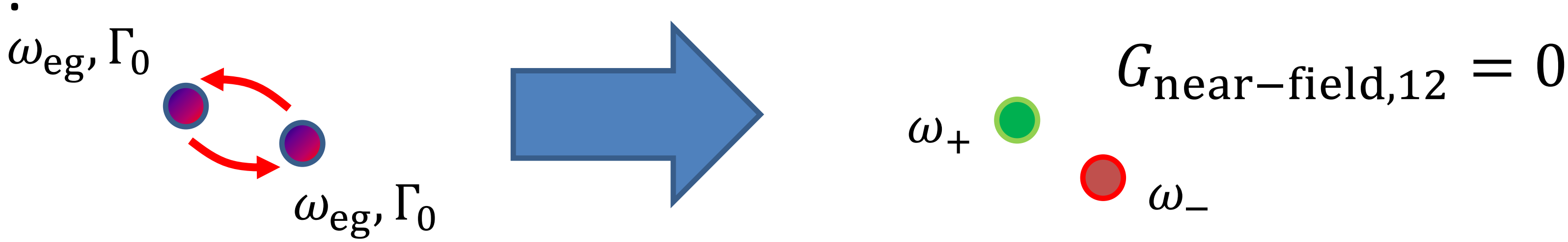


"Strong disorder renormalization group (RG)"

- Statistically, some pairs are extremely close to each other
  - Interact with each other much more strongly ($\sim 1/r^3$) than with all other atoms combined

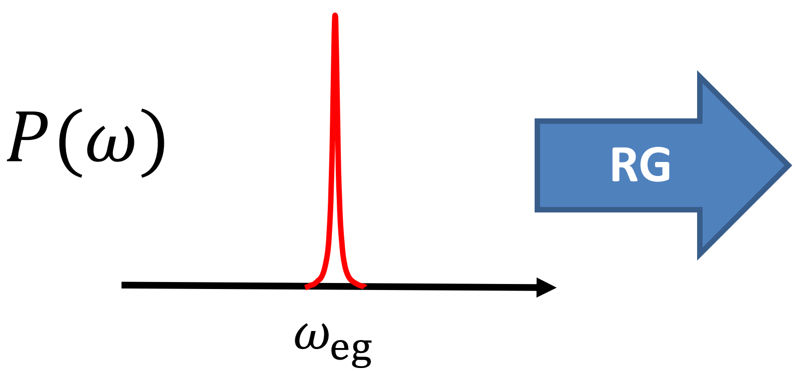
$$G^{(N\times N)} = G^{(2\times 2)}_{\text{near-field,pair}} + \left(G - G_{\text{near-field,pair}}\right)^{(N\times N)}$$

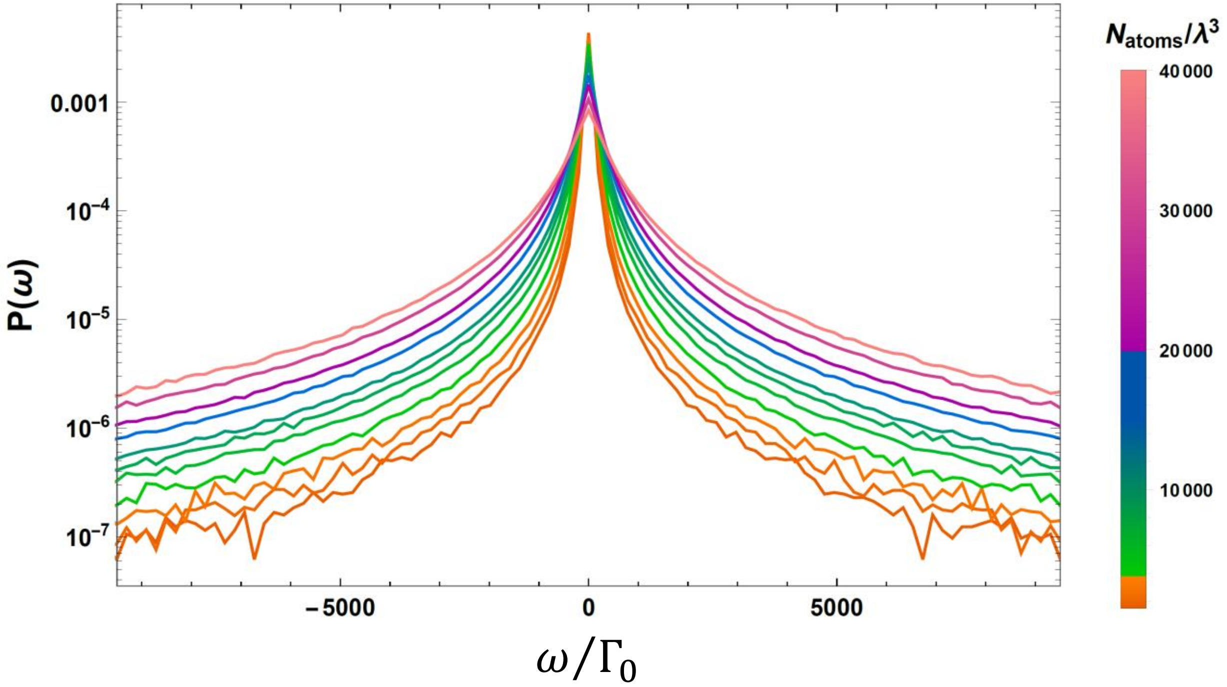
- Diagonalize these pairs first, replace with non-interacting atoms!



Many atoms: renormalization group

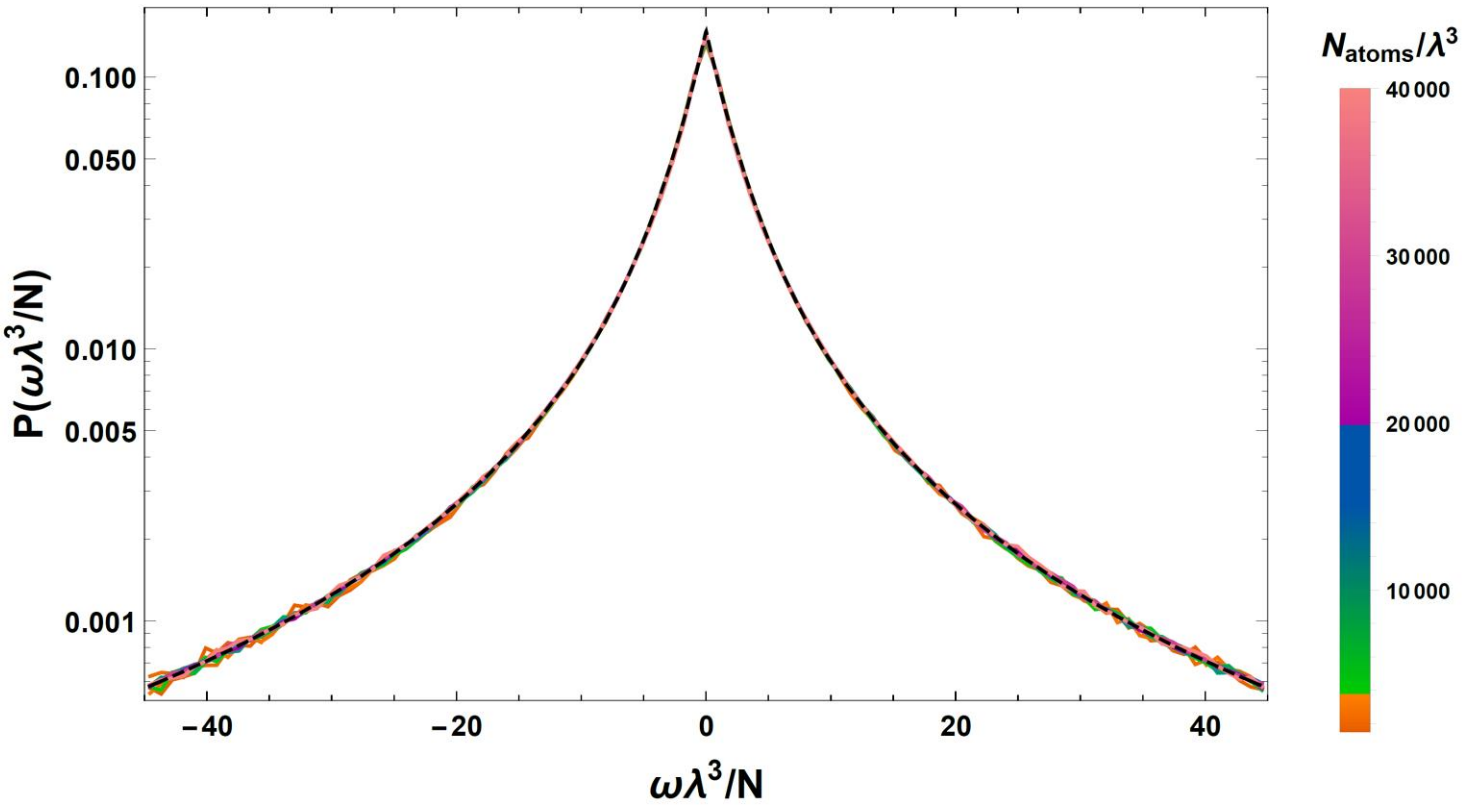
- Eliminate strong near-field interactions, leaving only far-field interactions
- Homogeneous atomic medium is optically **equivalent** to inhomogeneous medium with resonance distribution $P(\omega)$





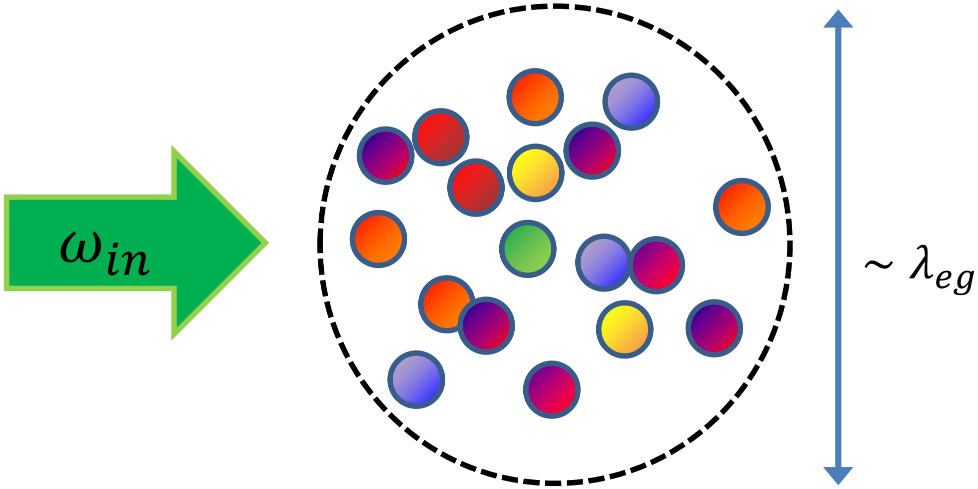
Origin of refractive index saturation

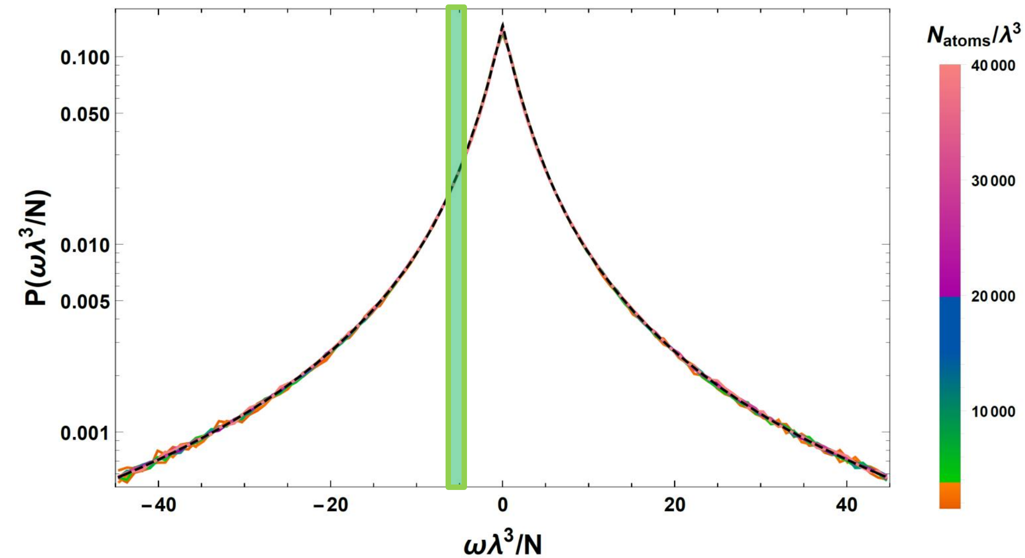
- Scale invariance when frequency is rescaled by density, $\omega \rightarrow \omega\lambda^3/N_{\text{atoms}}$



- Universal behavior!
  - "**Fixed point**" of RG flow
- Amount of broadening directly increases with density

Origin of refractive index saturation

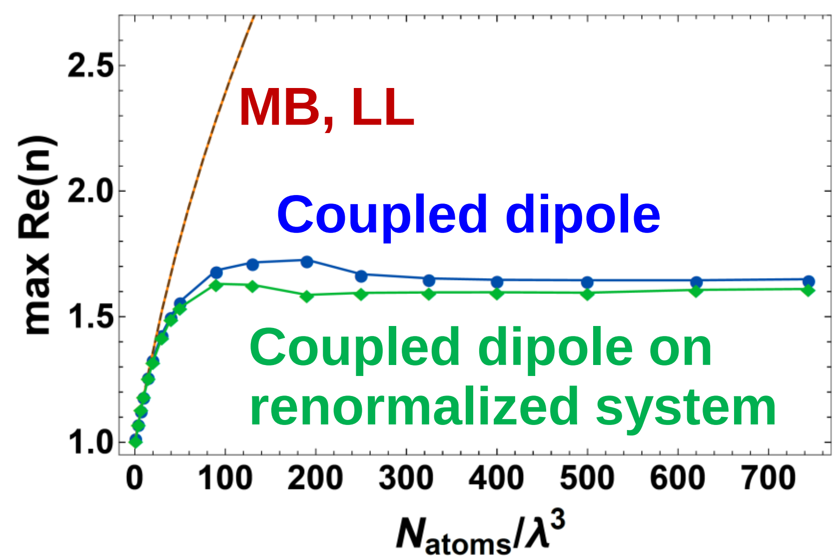




Renormalized ensemble

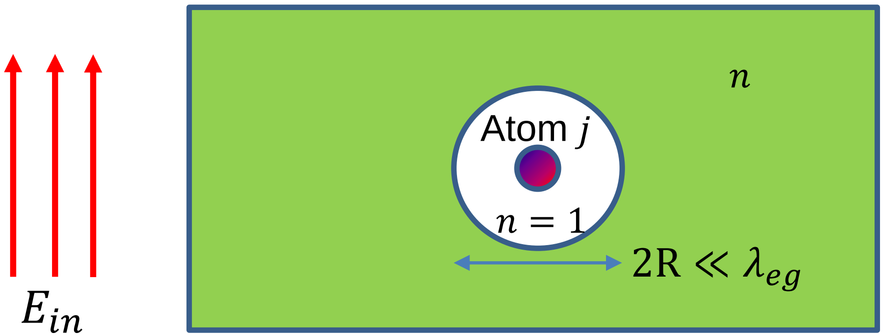
- At most ~ 1 near-resonant atom per cubic wavelength, per bandwidth $\sim \Gamma_0$, **regardless of physical density**

- Full simulations vs. RG:



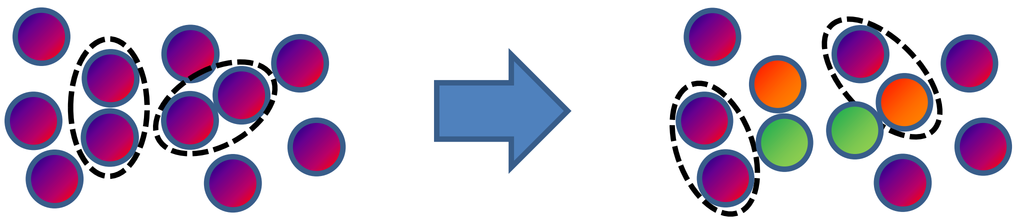
Why do conventional theories fail?

- Want to ignore granularity $\longrightarrow$ need to make **smooth medium** approximation
- Example: Lorentz-Lorenz model



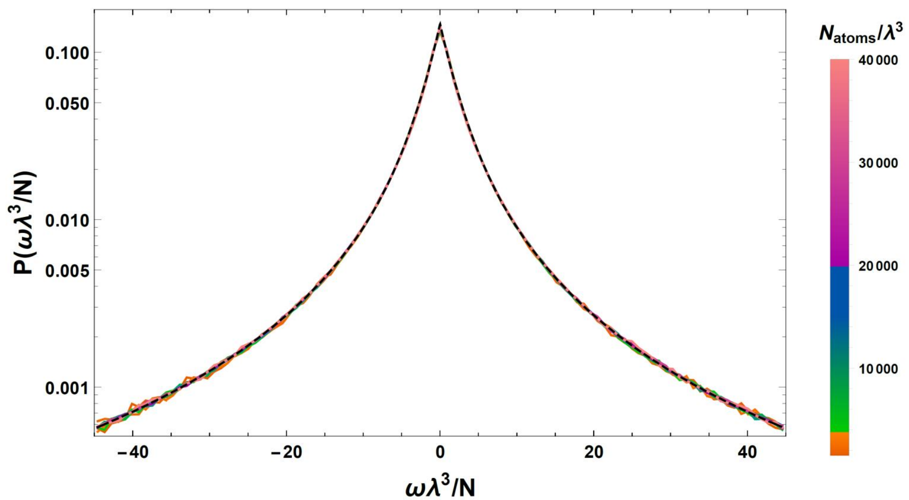
$$p_j \approx \alpha(\omega)\left[E_{in}(r_j,\omega) + \frac{n(\omega)^2 - 1}{3} E_{in}(r_j,\omega)\right]$$

- Renormalization group: **single nearest neighbors** matter most, due to $\sim 1/r^3$ interaction



A proper smooth medium theory

- Use RG to get rid of strong near-field interactions

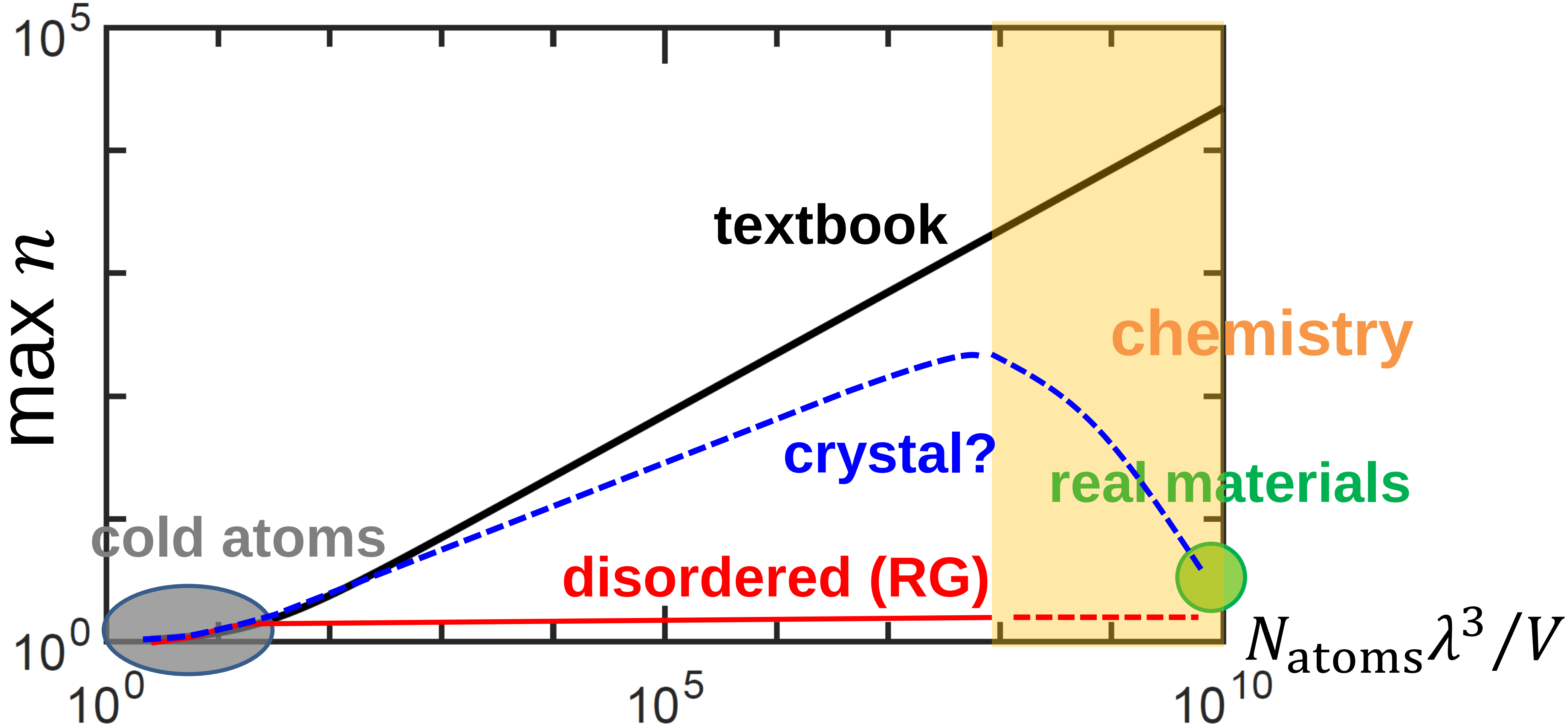


- Smooth medium approximation now valid!
  - Apply M-B equations to broadened medium $P(\omega_{\text{eff}})$:

$$n_{\text{MB}}(\omega)^2 \sim 1 + \int d\omega_{\text{eff}}\, \alpha(\omega - \omega_{\text{eff}}) P(\omega_{\text{eff}}) \frac{N_{\text{atoms}}}{V}$$

$$\max n \approx 1.8$$

Towards a complete theory of index?

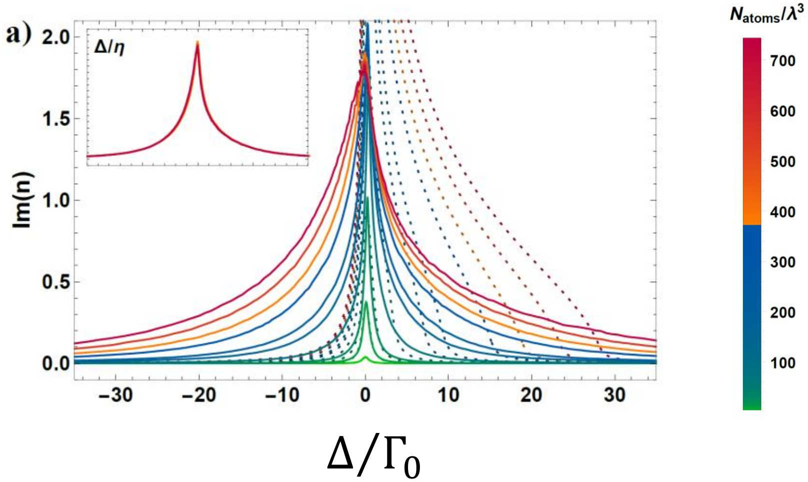


- Combined theory of multiple scattering and quantum chemistry?
- Short- or long-range atomic order?

Outlook: other possible implications

- RG as a general tool for multiple scattering in random media?
  - Nonlinear and quantum regimes?

- Quantum technologies based on atom-light interfaces



- Im(n) also reaches a limiting value

$$D \sim \mathrm{Im}(n) k_0 L$$ **Optical depth**

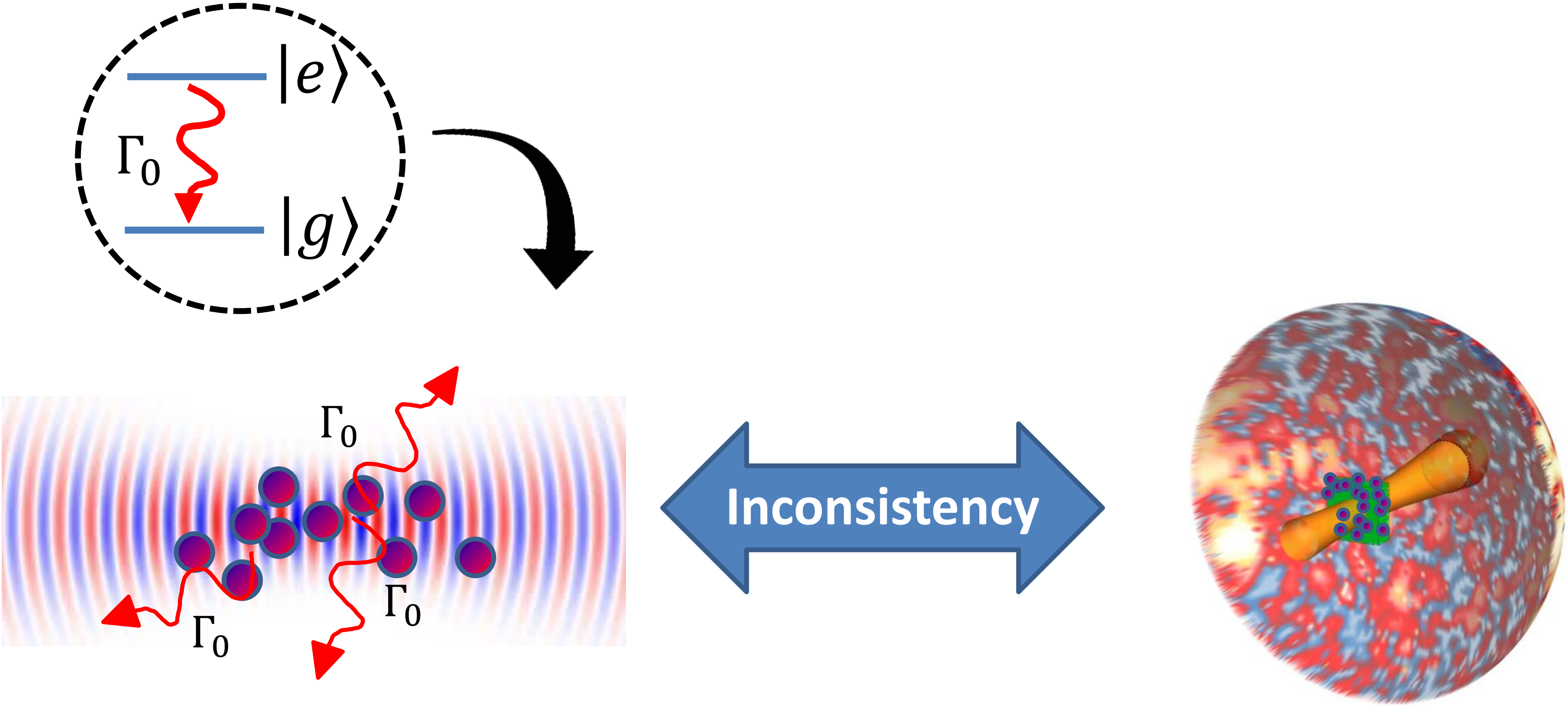
- D sets fundamental error bounds on almost every application

$\mathrm{Error} \sim 5.8/D$ for a quantum memory PRL 98, 123601 (2007)

- Bounds on minimum system size and maximum atomic density for high-fidelity quantum technologies?

Outlook: a quantum theory

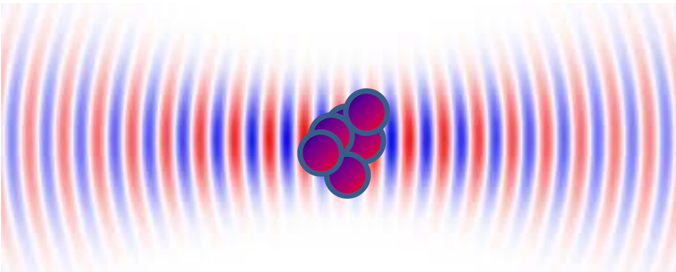
- Error bounds for applications are derived from quantum Maxwell-Bloch equations
  - Do not include interference and multiple scattering!



- Energy conservation $\rightarrow$ emission process must be correlated

Outlook: a quantum theory

- Famous historical example:

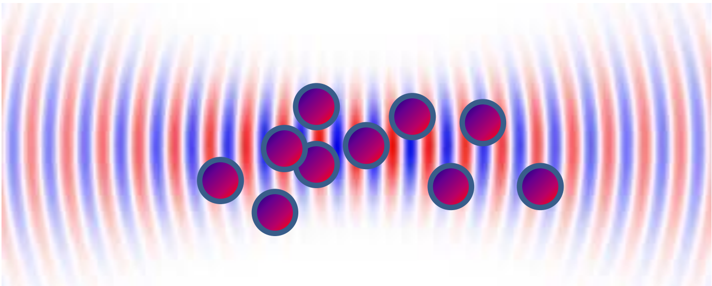


Dicke superradiance

Exactly solvable by collective spins

$$\hat{S} = \sum_j \sigma_{ge}^j$$

???



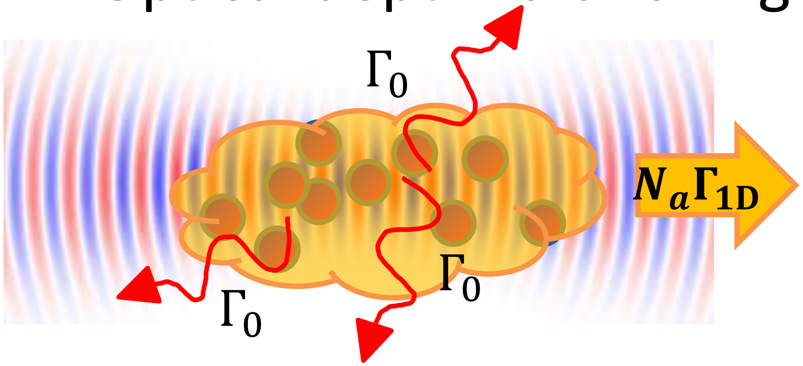
No symmetries

- One possible approach:

	Classical	**Quantum**
Degrees of freedom:	Dipoles p_j	"Spins" $\|g_j\rangle, \|e_j\rangle$
Total field:	$E(r) = E_{in}(r) + \sum_j G(r, r_j)\, p_j$	$\hat{E}(r) = \hat{E}_{\text{in}}(r) + \sum_j G(r, r_j)\, \hat{\sigma}_{ge}^j$
Dynamics:	$p_i = E_{\text{in}}(r_i) + \sum_j G(r_i, r_j)\, p_j$	$H = -\sum_{i,j} G(r_i, r_j) \sigma_{eg}^i \sigma_{ge}^j$

Outlook: a quantum theory

- Can we use interference as a **resource** in applications?
- Optical depth: branching ratio of information

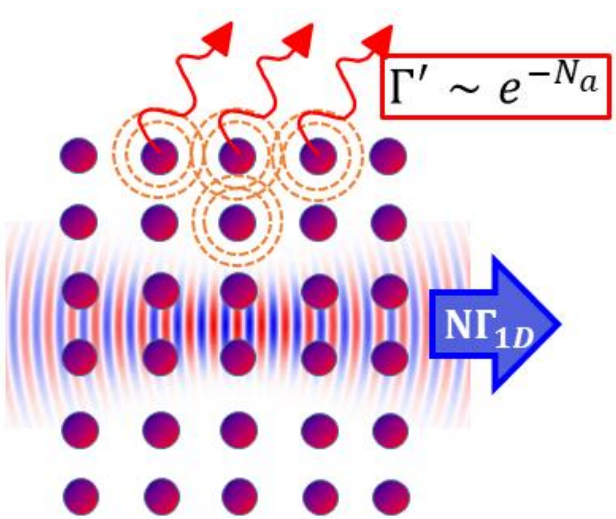


$$\boxed{D \sim \frac{N_a \Gamma_{1D}}{\Gamma_0}}$$

A.A. Svidzinsky et al, PRA 81, 053821 (2010)

Single collective excitation $|\psi\rangle \sim \sum_j e^{ik\cdot r_j}|e_j\rangle$

- Exploiting interference:



$$\boxed{\text{Branching ratio} \sim \frac{N\Gamma_{1D}}{\Gamma'(N)} \gg D}$$

- Quantum memory: Error $\sim e^{-D}$

A. Asenjo Garcia et al, PRX 7, 031024 (2017)

Summary

- Topic of light-matter interactions including multiple scattering is a rich and exciting frontier!