# Resonant dipole-dipole interaction in electromagnetically induced transparency

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QST, 25-27 August 2022

# Acknowledgements

Kuldeep Suthar, Postdoc
R. Ravisankar postdoc
Wei-Seng Hiew, NTU Master
Chung-Hsien Wang, NTU Master
Tzu-Hsuan Chang, Research assistant
Yi-Cheng Wang, Research assistant
Nai-Yu Tsai, NTU College project

Yu-Eng Wong, NTU College project till June 2022
Chun-Che Wang, Research assistant till June 2022
Chi-Chih Chen, Research assistant till March 2022
Yi-Chun Lin, NSYSU College project, till May 2022
Chin-Yang Lin, Research assistant, till March 2021

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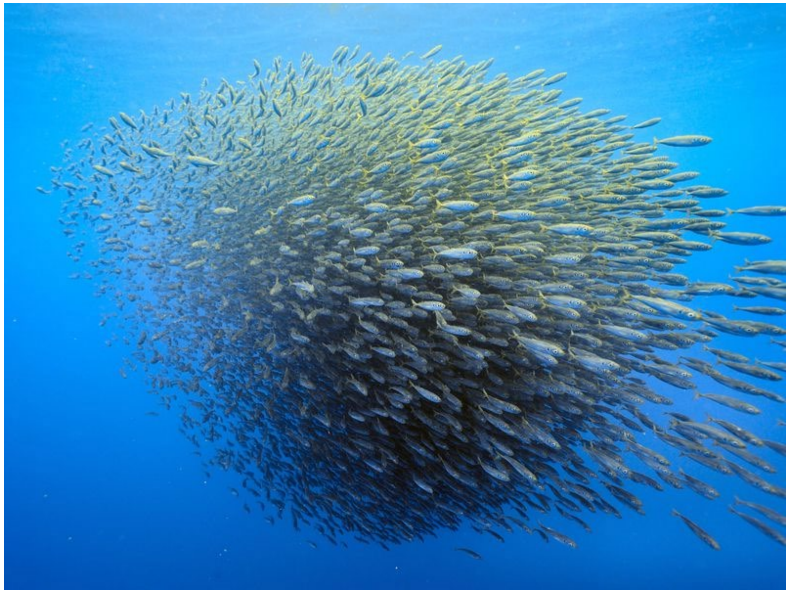
NCTS

# Outline

- Intro: Resonant dipole-dipole interactions
- EIT
- Perturbation treatments
- Beyond local-field approximation
- Dilemma in numerical simulations and Quantum Langevin Eqs

# Collectivity in nature



Complexity arises due to interactions (nonlinearity):

Fluctuations,

Multiple scales,

Emergent structure.

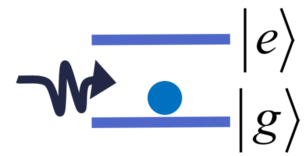
https://physics.aps.org/articles/v12/2

# Collectivity in atoms

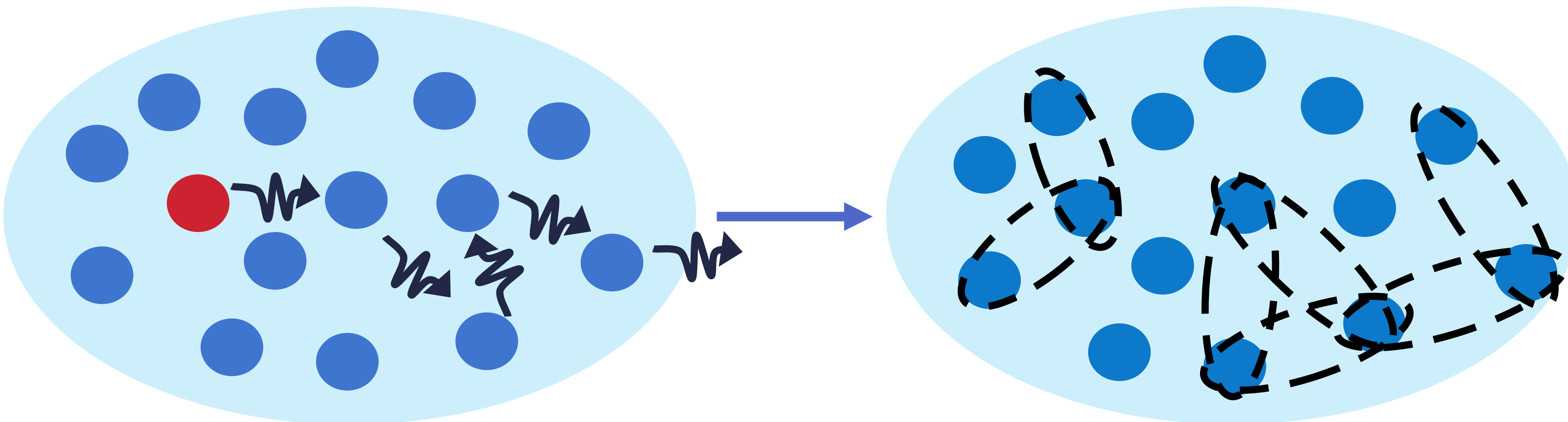
Complexity arises in nonlinear quantum systems as well:

Fluctuations, Multiple scales,

Emergent structure.

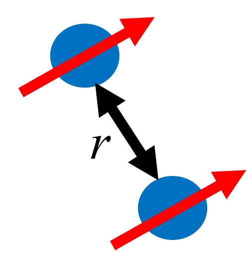



- ***Rescattering of light.***
- ***Dipole-dipole interactions.***

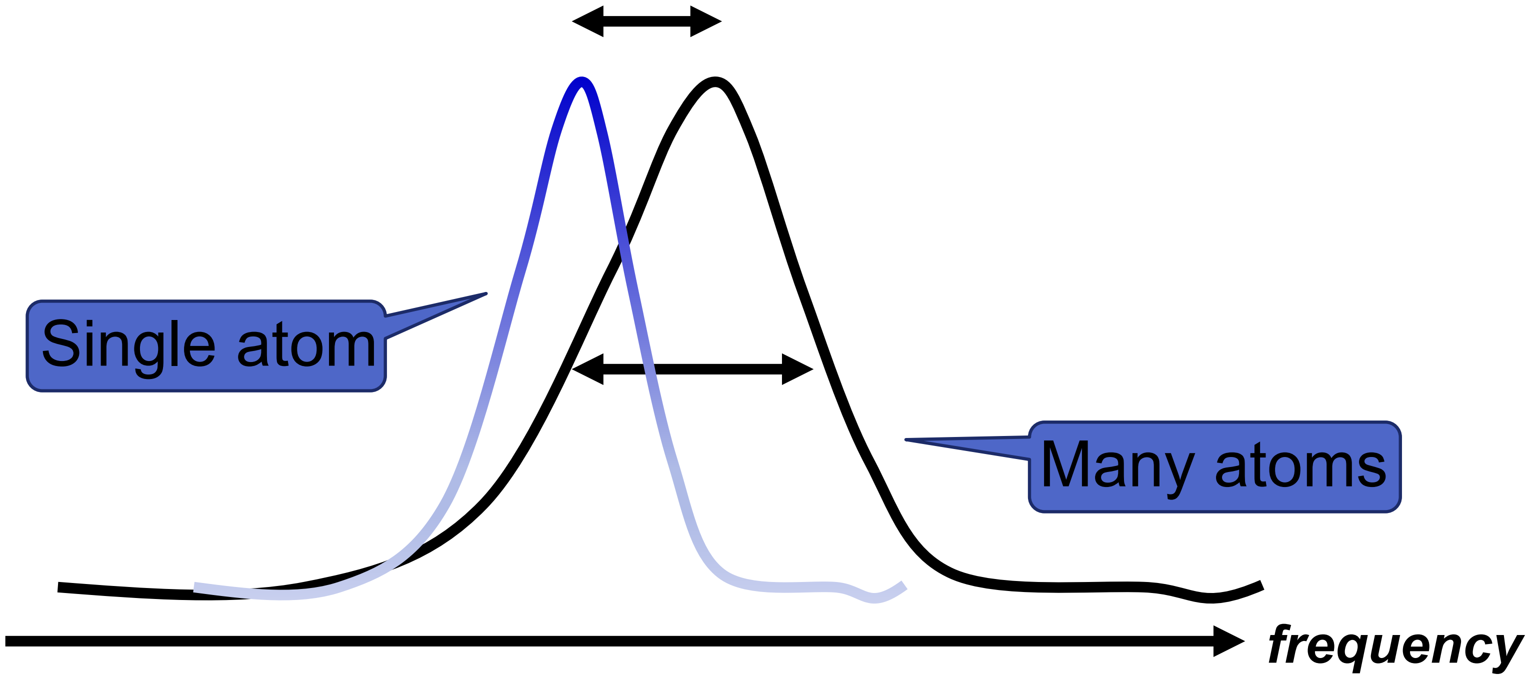


$$\text{RDDI}: V_{\alpha\beta} = -\frac{3\Gamma}{4(kr)^3}\left[p_{\alpha\beta}(ikr-1)+q_{\alpha\beta}(kr)^2\right]e^{ikr}.$$

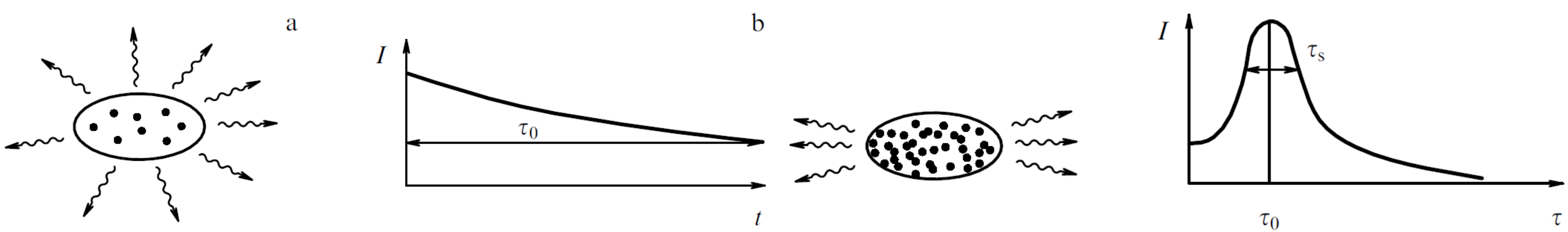
$$p_{\alpha\beta} \equiv \delta_{\alpha\beta} - (\hat{e}_\alpha^* \bullet \hat{r})(\hat{e}_\beta^* \bullet \hat{r}), q_{\alpha\beta} \equiv \delta_{\alpha\beta} - 3(\hat{e}_\alpha^* \bullet \hat{r})(\hat{e}_\beta^* \bullet \hat{r}).$$



# Collective effect in emission spectrum

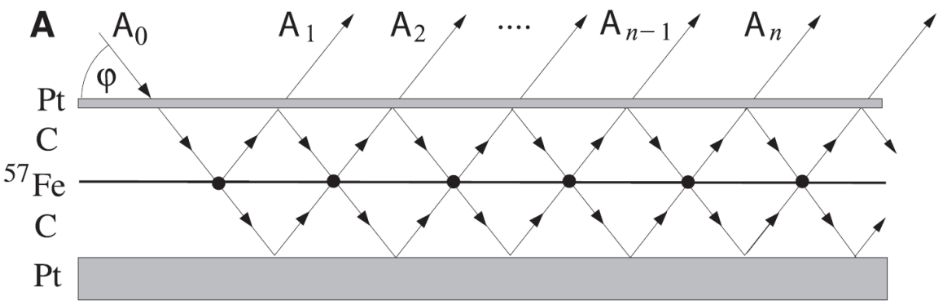



- ***Energy/frequency shift – Cooperative Lamb shift***
- ***Linewidth broadening***



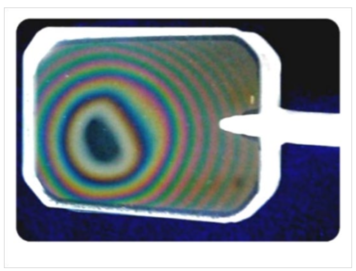

L. I. Men'shikov, Uspekhi 42 (2) 107 (1999).

## (1) Planar cavity:




R. Röhlsberger, et. al., Science 328, 1248 (2010).

## (2) Vapor cell:

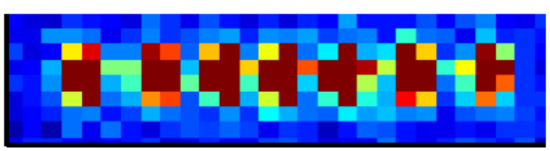


J. Keaveney, et. al., PRL 108, 173601 (2012)

## (3) Molecules:

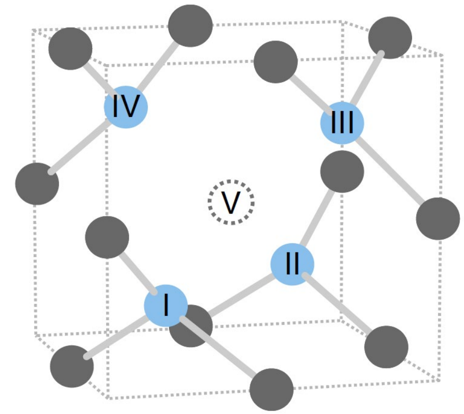
B. H. McGuyer, et. al., Nat. Phys. 11, 32 (2014).

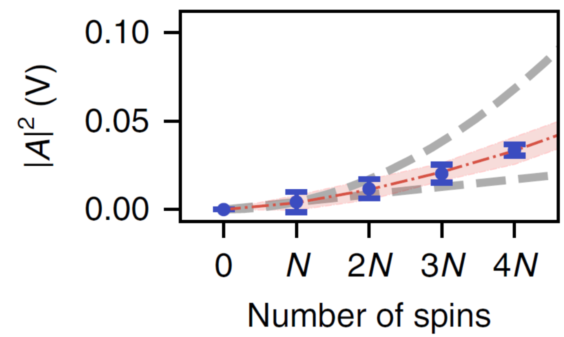
## (4) Ion chains:



Z. Meir, et. al., PRL 113, 193002 (2014)

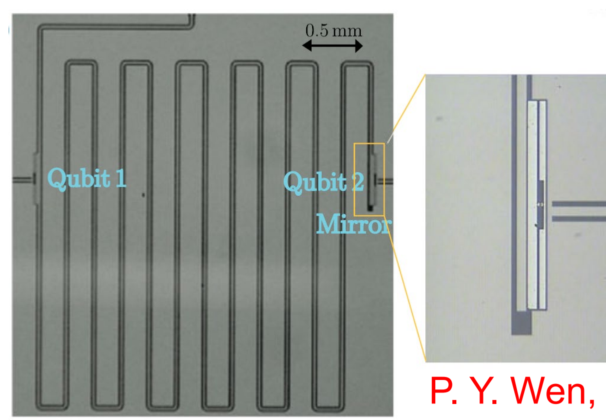
## (5) NV centers in diamond:



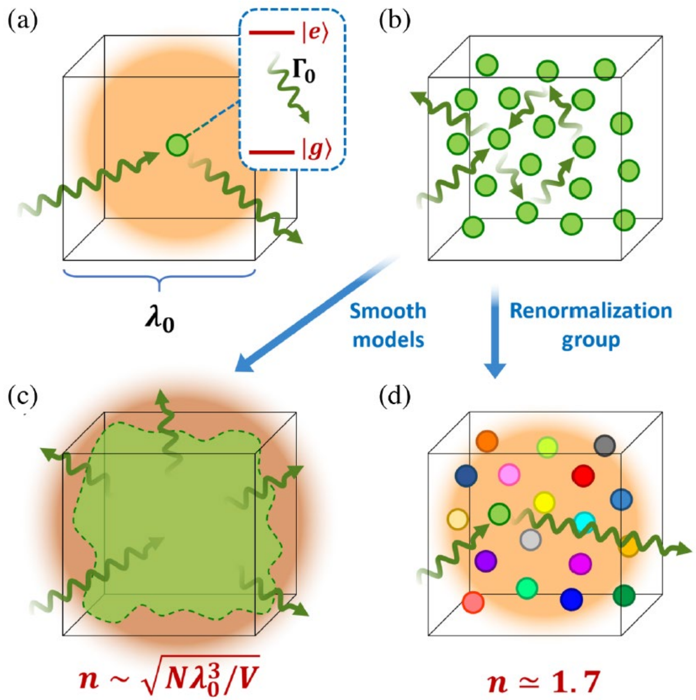

A. Angerer, et. al., Nat. Phys.14, 1168 (2018)

## (6) Superconducting qubits:

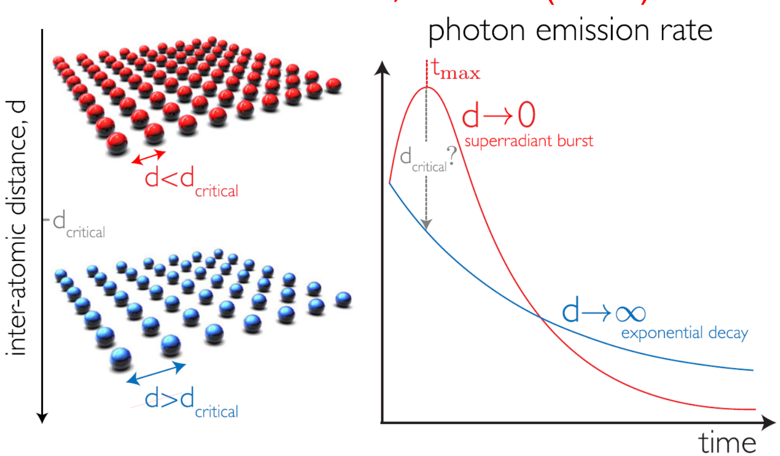



P. Y. Wen, et. al., PRL 123, 233602 (2019)

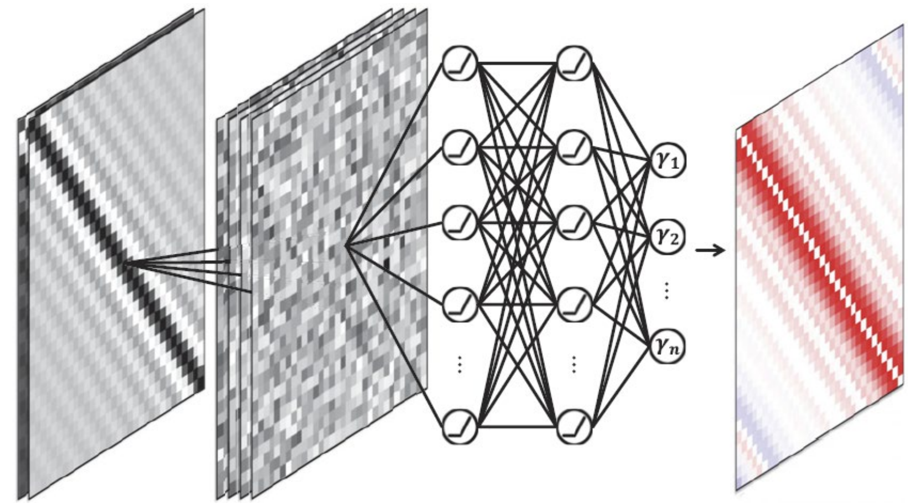
# Recent theoretical attempts

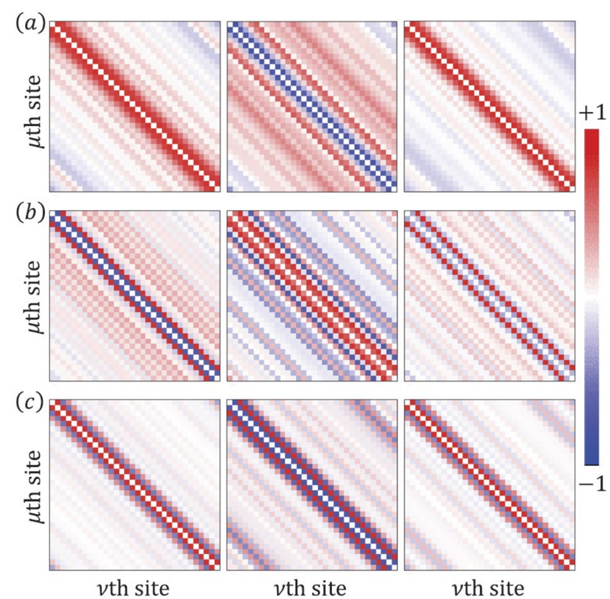



PRX 11, 011026 (2021)




NATURE COMMUN. (2022) 13:2285




JPB 55, 135501 (2022)

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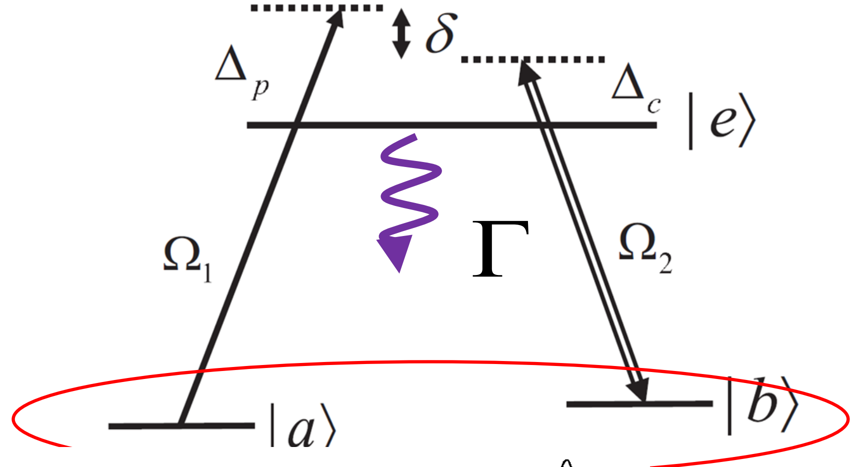
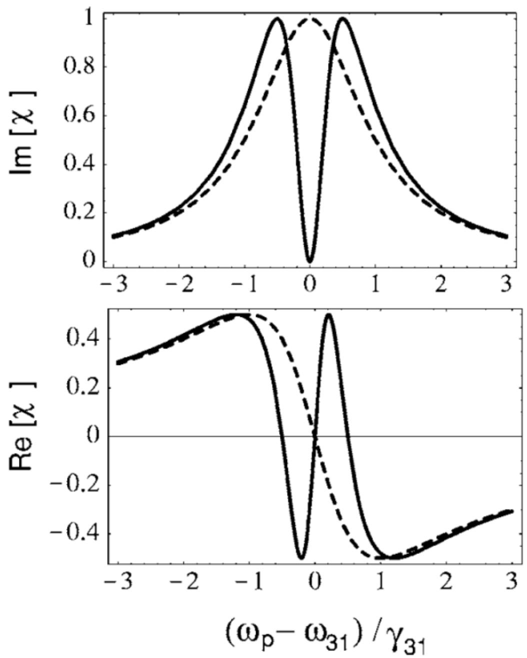
- EIT
- Perturbation treatments
- Beyond local-field approximation
- Dilemma in numerical simulations and Quantum Langevin Eqs

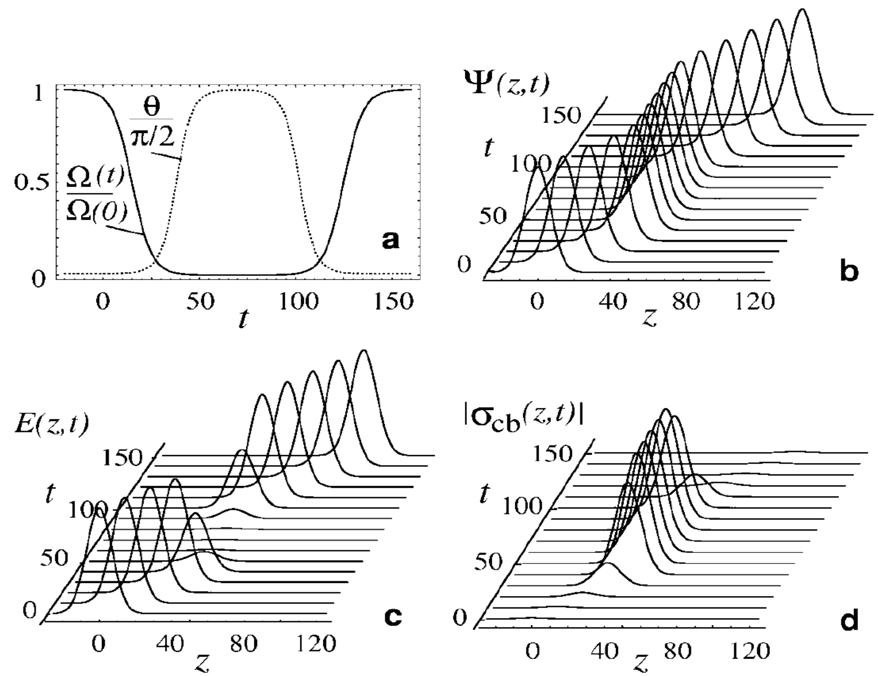
# EIT configuration

The appearance of the second (coupling) field enables quantum interference, making the light propagation highly nontrivial.

$$\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=\frac{ik}{2\varepsilon_0}P(z,t)$$




M. Fleischhauer et al, RMP **77**, 633 (2000); M. Fleischhauer and M. D. Lukin, PRL **84**, 5094 (2000)

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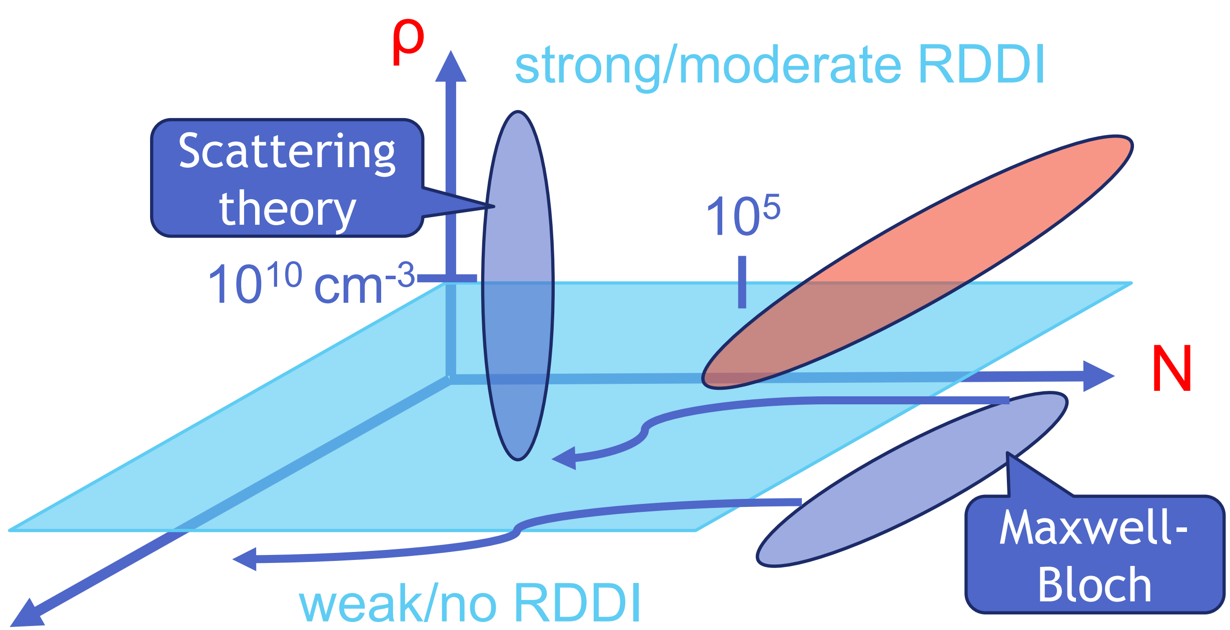
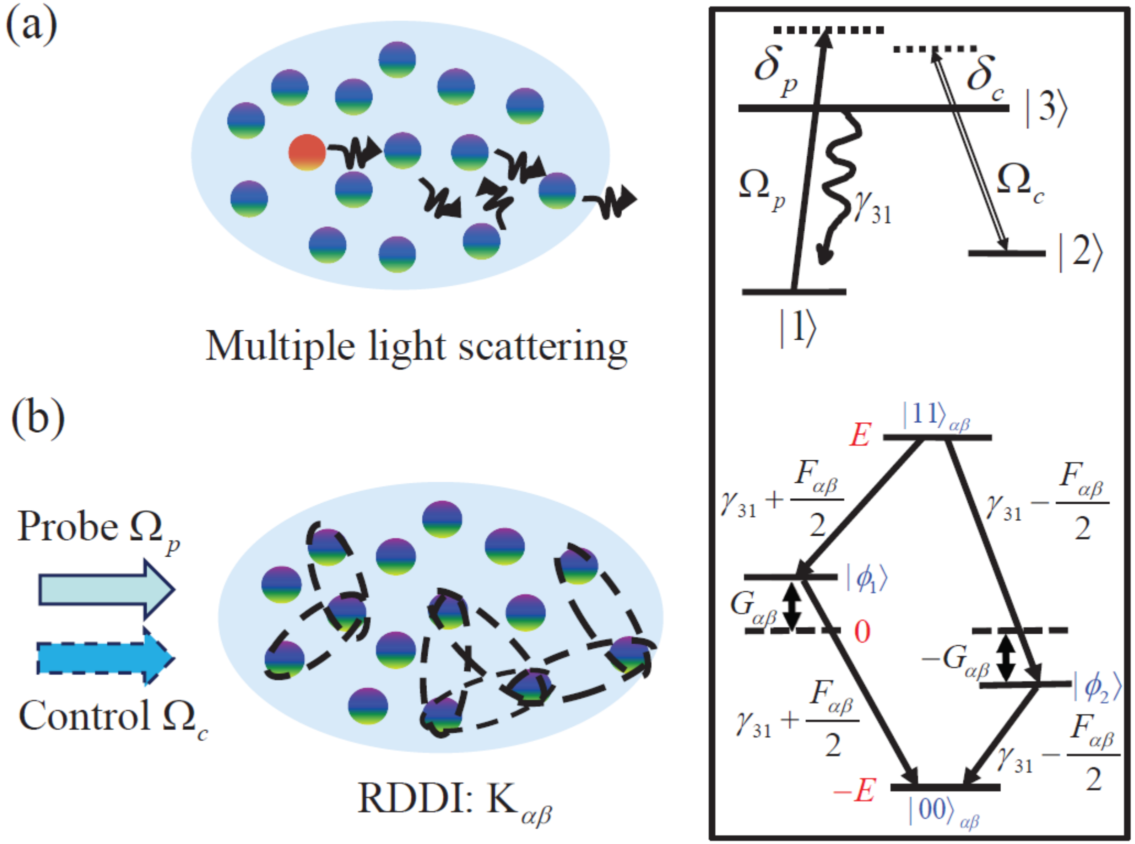
# Dilemma

parameters are $N \sim 5 \times 10^9$ in a cloud size of $3 \times 3 \times 14$ mm$^3$, which has an average atomic density $\sim 4 \times 10^{10}$ cm$^{-3}$. For a peak density around $10^{11}$ cm$^{-3}$ and considering an interaction volume determined by probe field propagation along the long axis with a cylindrical geometry of $\pi(0.2)^2 \times 14$ mm$^3$, we have $N \sim 1.8 \times 10^8$ with an optical density $\sim 400$.

(a) State of system.
(b) System parameters.



# RDDI in EIT




- Neglect of RDDI in control field transition.
- Two-level diagram demonstration.

PRA 105, 063711 (2022)

# RDDI in EIT: Hamiltonian

$$\hat{H}_I = \hbar\delta_p \sum_{\mu=1}^{N} \hat{\sigma}_{11}^{\mu} + \hbar\delta_c \sum_{\mu=1}^{N} \hat{\sigma}_{22}^{\mu} + \sum_{\mu=1}^{N} \left( -\frac{\hbar\Omega_p}{2} e^{i\mathbf{k}_p\cdot\mathbf{r}_\mu} \hat{\sigma}_{13}^{\mu,\dagger} + h.c. \right) + \sum_{\mu=1}^{N} \left( -\frac{\hbar\Omega_c}{2} e^{i\mathbf{k}_c\cdot\mathbf{r}_\mu} \hat{\sigma}_{23}^{\mu,\dagger} + h.c. \right)$$

$$\frac{d\hat{Q}}{dt} = -\frac{i}{\hbar}[\hat{Q}, \hat{H}_I] - i \sum_{\mu\neq\nu,\nu}^{N} [\hat{Q}, G_{\mu\nu}\hat{\sigma}_{31}^{\mu}\hat{\sigma}_{13}^{\nu}] + \mathcal{L}_p[\hat{Q}] + \mathcal{L}_c[\hat{Q}] + \mathcal{L}_g[\hat{Q}],$$

Master
equations

$$\mathcal{L}_p[\hat{Q}] \equiv -\sum_{\mu,\nu}^{N} \frac{F_{\mu\nu}}{2} (\hat{\sigma}_{31}^{\mu}\hat{\sigma}_{13}^{\nu}\hat{Q} + \hat{Q}\hat{\sigma}_{31}^{\mu}\hat{\sigma}_{13}^{\nu} - 2\hat{\sigma}_{31}^{\mu}\hat{Q}\hat{\sigma}_{13}^{\nu})$$

Lindblad
forms

$$\mathcal{L}_c[\hat{Q}] \equiv -\sum_{\mu,\nu}^{N} \gamma_{32} (\hat{\sigma}_{32}^{\mu}\hat{\sigma}_{23}^{\nu}\hat{Q} + \hat{Q}\hat{\sigma}_{32}^{\mu}\hat{\sigma}_{23}^{\nu} - 2\hat{\sigma}_{32}^{\mu}\hat{Q}\hat{\sigma}_{23}^{\nu}),$$

$$\mathcal{L}_g[\hat{Q}] \equiv -\sum_{\mu,\nu}^{N} \gamma_{21} (\hat{\sigma}_{21}^{\mu}\hat{\sigma}_{12}^{\nu}\hat{Q} + \hat{Q}\hat{\sigma}_{21}^{\mu}\hat{\sigma}_{12}^{\nu} - 2\hat{\sigma}_{21}^{\mu}\hat{Q}\hat{\sigma}_{12}^{\nu}).$$

# RDDI in EIT: EOM

RDDI effect

$$\frac{d}{d\tau}\tilde{\sigma}_{23} = (i\delta_c - \gamma_{21} - \gamma_{32})\tilde{\sigma}_{23} + i\frac{\Omega_c}{2}(\tilde{\sigma}_{22} - \tilde{\sigma}_{33}) + i\frac{\Omega_p}{2}\tilde{\sigma}^{\dagger}_{12},$$

$$\frac{d}{d\tau}\tilde{\sigma}_{13} = (i\delta_p - \gamma_{31})\tilde{\sigma}_{13} + i\frac{\Omega_c}{2}\tilde{\sigma}_{12} + i\frac{\Omega_p}{2}(\tilde{\sigma}_{11} - \tilde{\sigma}_{33}) - \frac{1}{N_z}\sum_{\alpha=1}^{N_z}\sum_{\beta\neq\alpha}^{N} K_{\alpha\beta}(\tilde{\sigma}^{\alpha}_{11} - \tilde{\sigma}^{\alpha}_{33})\tilde{\sigma}^{\beta}_{13},$$

$$\frac{d}{d\tau}\tilde{\sigma}_{12} = (i\delta_2 - \gamma_{21})\tilde{\sigma}_{12} - i\frac{\Omega_p}{2}\tilde{\sigma}^{\dagger}_{23} + i\frac{\Omega_c^*}{2}\tilde{\sigma}_{13},$$

$$\frac{d}{d\tau}\tilde{\sigma}_{11} = 2\gamma_{31}\tilde{\sigma}_{33} + 2\gamma_{21}\tilde{\sigma}_{22} - i\frac{\Omega_p}{2}\tilde{\sigma}^{\dagger}_{13} + i\frac{\Omega_p^*}{2}\tilde{\sigma}_{13} + \frac{1}{N_z}\sum_{\alpha=1}^{N_z}\sum_{\beta\neq\alpha}^{N}\left[K_{\alpha\beta}(\tilde{\sigma}^{\alpha}_{31}\tilde{\sigma}^{\beta}_{13}) + K^*_{\alpha\beta}(\tilde{\sigma}^{\beta}_{31}\tilde{\sigma}^{\alpha}_{13})\right],$$

$$\frac{d}{d\tau}\tilde{\sigma}_{33} = -\gamma_3\tilde{\sigma}_{33} + i\frac{\Omega_p}{2}\tilde{\sigma}^{\dagger}_{13} - i\frac{\Omega_p^*}{2}\tilde{\sigma}_{13} + i\frac{\Omega_c}{2}\tilde{\sigma}^{\dagger}_{23} - i\frac{\Omega_c^*}{2}\tilde{\sigma}_{23} - \frac{1}{N_z}\sum_{\alpha=1}^{N_z}\sum_{\beta\neq\alpha}^{N}\left[K_{\alpha\beta}(\tilde{\sigma}^{\alpha}_{31}\tilde{\sigma}^{\beta}_{13}) + K^*_{\alpha\beta}(\tilde{\sigma}^{\beta}_{31}\tilde{\sigma}^{\alpha}_{13})\right],$$

$$\frac{d}{dz}\Omega_p = \frac{iD\Gamma}{2L}\tilde{\sigma}_{13},$$

- Only first-order cumulant kept, coarse-grained and slow-varying treatments.
- Neglect of quantum noises and RDDI emerges from system-reservoir interaction.
- Same Maxwell-Bloch Eq. and valid from the side of noninteracting regime.

# RDDI in EIT: Perturbative treatment

$$\frac{d}{d\tau}\tilde{\sigma}_{12} \approx (i\delta_2 - \gamma_{21})\tilde{\sigma}_{12} + i\frac{\Omega_c^*}{2}\tilde{\sigma}_{13},$$

$$\frac{d}{d\tau}\tilde{\sigma}_{13} \approx (i\delta_p - \gamma_{31})\tilde{\sigma}_{13} + i\frac{\Omega_c}{2}\tilde{\sigma}_{12} + i\frac{\Omega_p}{2}$$

$$-\frac{1}{N_z}\sum_{\alpha=1}^{N_z}\sum_{\beta\neq\alpha}^{N} K_{\alpha\beta}\tilde{\sigma}_{13}^{\beta}.$$

Weak-field approx.

$$\tilde{\sigma}_{12} = \frac{-\Omega_c^*/2}{\delta_2 + i\gamma_{21}}\tilde{\sigma}_{13}$$

Steady-state

$$\tilde{\sigma}_{13}^{(0)}(z) = \frac{-i\Omega_p(z)/2}{i\delta_p - \gamma_{31} - \frac{i\Omega_c^2/4}{\delta_2 + i\gamma_{21}}} \equiv \frac{-i\Omega_p(z)/2}{A}$$

$$\tilde{\sigma}_{13}^{(1)}(z) = \frac{-i\Omega_p(z)/2}{A} + \frac{1}{AN_z}\sum_{\alpha=1}^{N_z}\sum_{\beta\neq\alpha}^{N} K_{\alpha\beta}\tilde{\sigma}_{13}^{\beta,(0)}(\mathbf{r}_\beta)$$

First iteration.

# RDDI in EIT: Perturbative treatment

$$\tilde{\sigma}_{13}^{(1)}(z) = \frac{-i\Omega_p(z)/2}{A} + \frac{f_C}{A}\frac{-i\Omega_p(z)/2}{A}$$

$$T_M = \exp\left[\frac{D\Gamma}{2}\mathrm{Re}\left(\sum_{m=0}^{M}\frac{f_C^m}{A^{m+1}}\right)\right]$$

Up to Mth order scattering in transmission.

$$T = \exp\left[\frac{D\Gamma}{2}\mathrm{Re}\left(\frac{1}{A - f_C}\right)\right]$$

After local-field approx.

$$A \equiv i\delta_p - \gamma_{31} - \frac{i\Omega_c^2/4}{\delta_2 + i\gamma_{21}},$$

$$f_C \equiv \frac{1}{N_z}\sum_{\alpha=1}^{N_z}\sum_{\beta\neq\alpha}^{N} K_{\alpha\beta}.$$

Collective frequency shift and decay rate.

$$\tilde{\delta}_p = \delta_p - \mathrm{Im}[f_C],\ \tilde{\gamma}_{31} = \gamma_{31} + \mathrm{Re}[f_C]$$

# RDDI in EIT: Results

$$\tilde{\gamma}_{31} = N\gamma_{31} \int_{-\infty}^{\infty} dxdydz(\pi^{3/2} R_{\perp}^{-2} R_L^{-1}) e^{-\frac{x^2+y^2}{R_{\perp}^2}} e^{-\frac{z^2}{R_L^2}}$$

$$\times \int \frac{3}{8\pi} \left(1 - \frac{\sin^2\theta}{2}\right) d\Omega_{\mathbf{k}} e^{-i\mathbf{k}_p\cdot(\mathbf{r}_\alpha - \mathbf{r})} e^{i\mathbf{k}\cdot(\mathbf{r}_\alpha - \mathbf{r})}$$

When $R_\perp = R_L$ and with $|\mathbf{k}_p|R_L \gg 1$, we can further simplify the above to

$$\tilde{\gamma}_{31} = \frac{3N\gamma_{31}}{8} \frac{2}{|\mathbf{k}_p|^2 R_L^2/2 + i|\mathbf{k}_p||\mathbf{r}_\alpha|\cos\theta'} \quad (19)$$

$$\approx \frac{3N\gamma_{31}}{2|\mathbf{k}_p|^2 R_L^2}. \quad (20)$$

Sphere-like

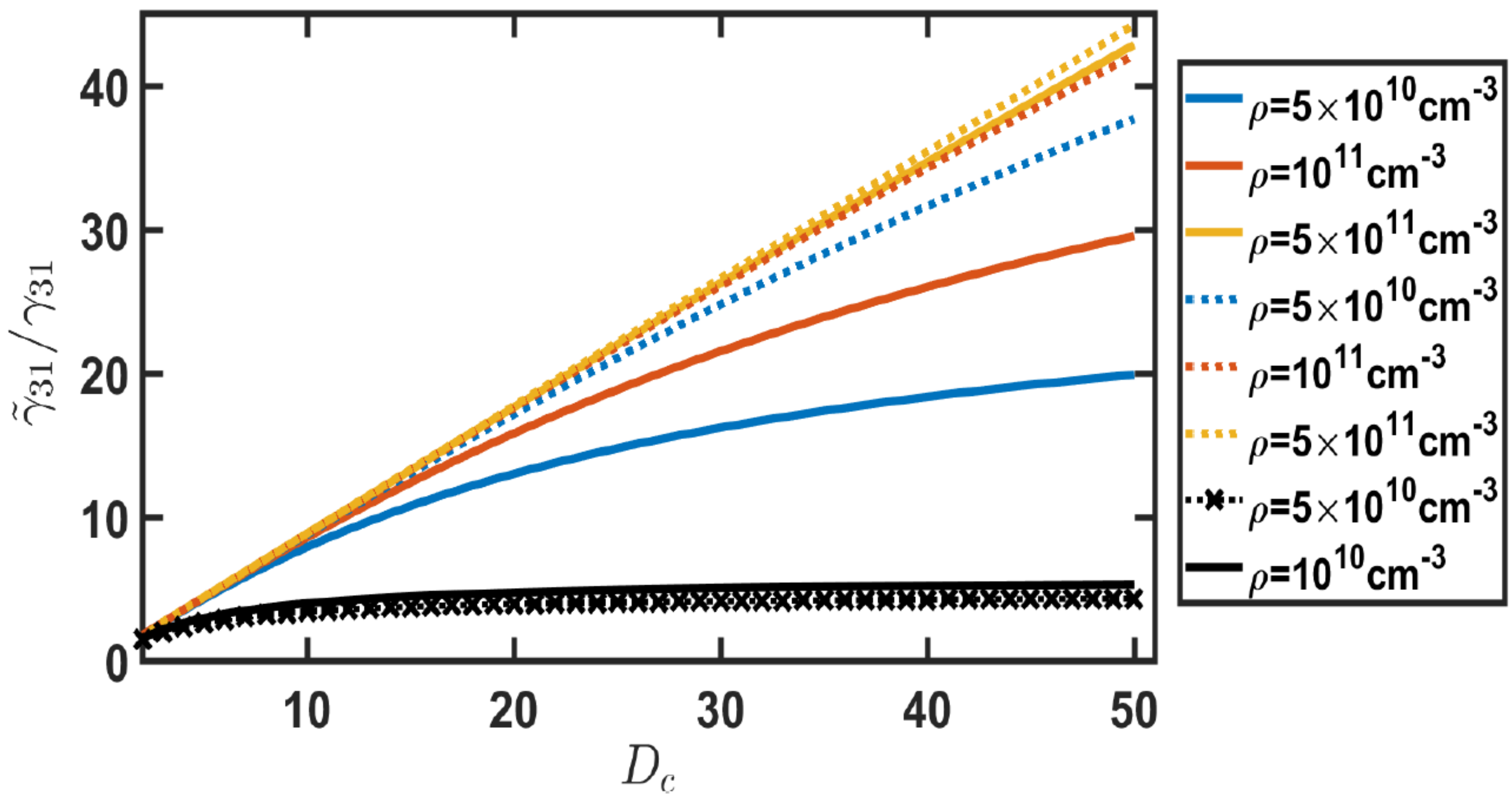
$$\tilde{\gamma}_{31}/\gamma_{31} = \frac{3N}{8} \frac{\sqrt{\pi}4am^2}{4a^{3/2}m^{5/2}} \times \frac{1}{2} = \frac{3\sqrt{\pi}}{8} \frac{N}{|\mathbf{k}_p|R_L}$$

$$\tilde{\gamma}_{31}/\gamma_{31} = \frac{3\sqrt{\pi}}{8} \frac{N}{|\mathbf{k}_p|R_L} = \frac{\pi}{8} D_c \frac{|\mathbf{k}_p|R_\perp^2}{2R_L}$$

Needle-like

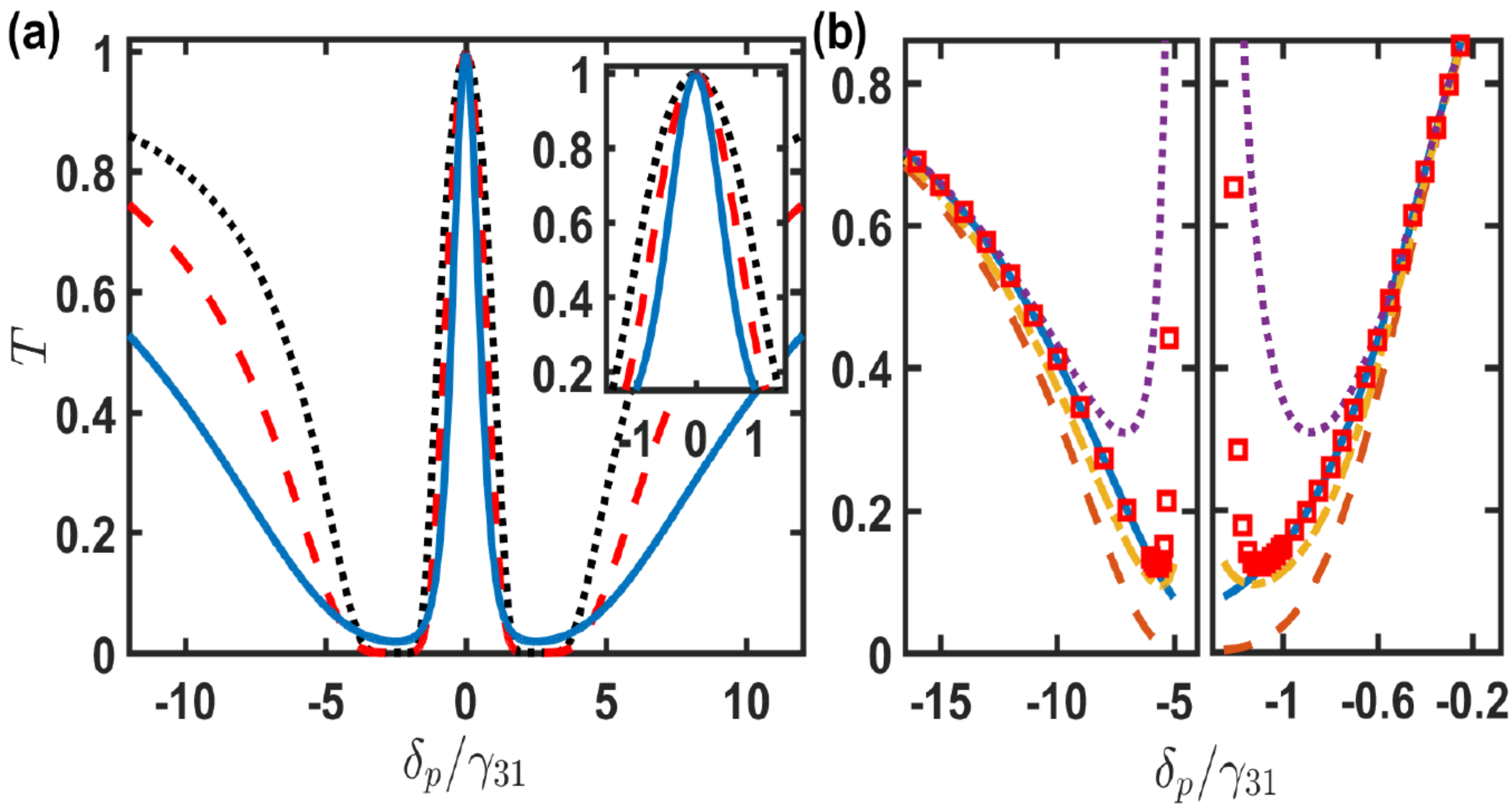
- Mutual dependence of N, Dc, R’s, and atomic density.

# RDDI in EIT: linewidth



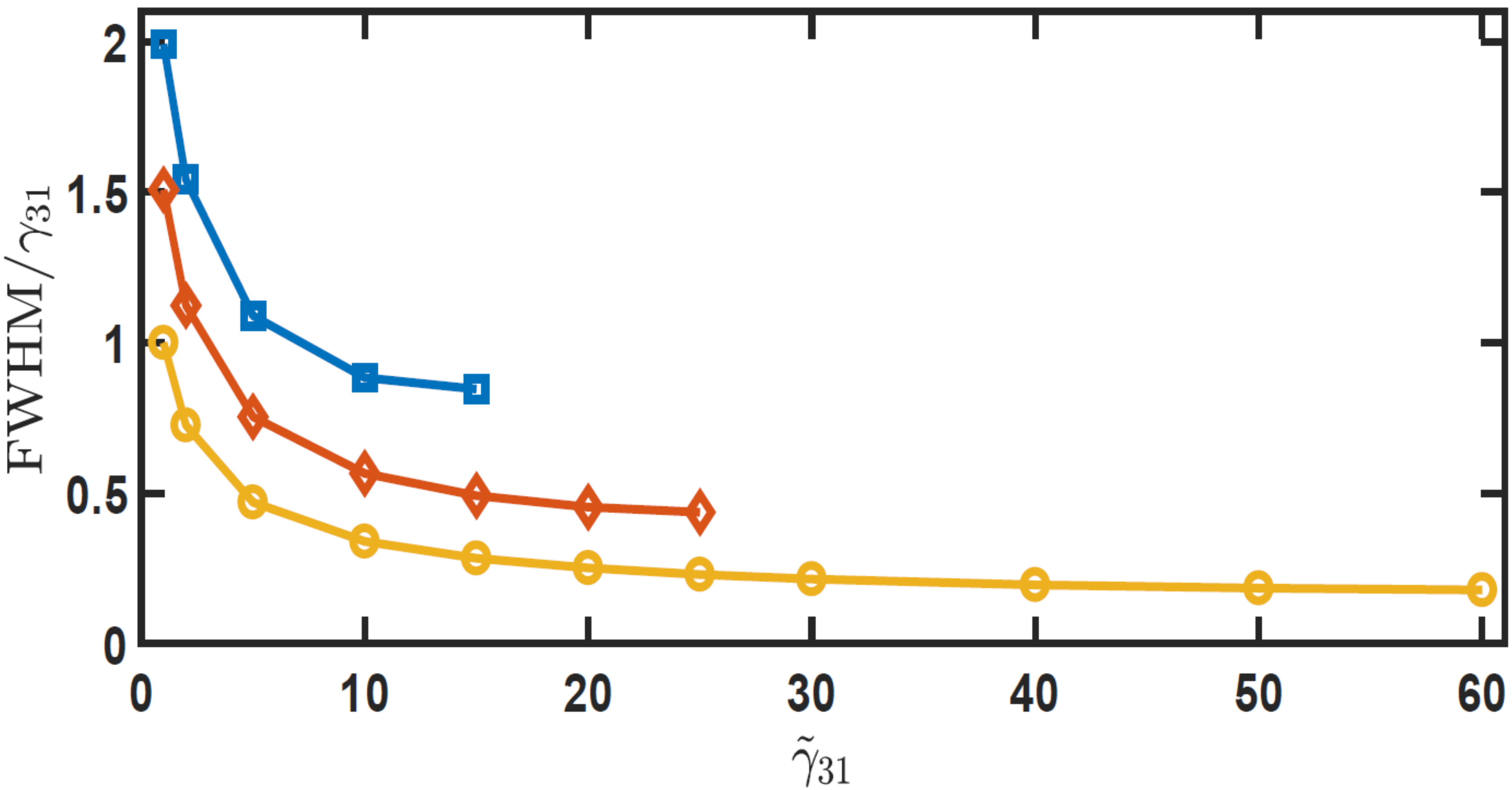

- Quite deviated range of predicted collective decay rates.

# RDDI in EIT: EIT spectrum




- Multiple scattering of RDDI and narrowing of transparency window.

# RDDI in EIT: FWHM




- narrowing of transparency window: storage efficiency reduced.

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# RDDI in EIT: Beyond local-field approx.

$$0 = \left[ i\delta_p - \gamma_{31} - \frac{i|\Omega_c|^2}{4(\delta_2 + i\gamma_{21})} \right] \tilde{\sigma}_{13}^{\alpha} + i\frac{\Omega_p(\mathbf{r}_\alpha)}{2}$$

$$- \sum_{\beta \neq \alpha}^{N} K_{\alpha\beta} \tilde{\sigma}_{13}^{\beta},$$

$$i\hat{M}\vec{\sigma}_{13} = -\frac{\vec{\Omega}_p}{2},$$

$$\vec{\sigma}_{13} = \frac{i}{2}\hat{M}^{-1}\vec{\Omega}_p$$

$$M = \begin{bmatrix} -A & K_{1,2} & K_{1,3} & \dots & K_{1,N} \\ K_{2,1} & -A & K_{2,3} & \dots & K_{2,N} \\ K_{3,1} & K_{3,2} & -A & \dots & \vdots \\ \vdots & \vdots & \vdots & \ddots & K_{N-1,N} \\ K_{N,1} & \dots & \dots & K_{N,N-1} & -A \end{bmatrix}$$

$$\left( -\frac{i}{2k_p}\nabla_\perp^2 + \frac{\partial}{\partial z} \right) \Omega_p(\mathbf{r}) = -\frac{D\Gamma}{4L} \int \hat{M}^{-1}(\mathbf{r} - \mathbf{r}')$$

$$\times \Omega_p(\mathbf{r}')\rho(\mathbf{r}')d\mathbf{r}',$$

$$\bar{\Omega}_p(s) = \frac{\Omega_p(z=0)}{s + \frac{D\Gamma}{4L}(N/L)\bar{M}^{-1}(s)}$$

Laplace transform

# RDDI in EIT: Beyond local-field approx.

Now a weak RDDI can be approximated by a far-field expression in $K(r-r') \to (3\Gamma/2)(-ie^{i\xi}e^{-i\mathbf{k}_p\cdot(\mathbf{r}-\mathbf{r}')})/\xi$. We finally obtain

$$\frac{\Omega_p(z=L)}{\Omega_p(z=0)} = \frac{1}{2\pi i}\oint \frac{e^{sL}ds}{s-\frac{D\Gamma}{4AL}-\frac{D\Gamma}{4AL}(\frac{N}{k_pL})(\frac{3\Gamma/2}{A})e^{as}\bar{\Gamma}(0,as)}, \quad (36)$$

where $a$ is introduced in Laplace transform of $1/(|r-r'|+a)$

- Multiple residues → multiple resonances and decay modes.

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# Dilemma and future direction

parameters are $N \sim 5 \times 10^9$ in a cloud size of $3 \times 3 \times 14$ mm$^3$, which has an average atomic density $\sim 4 \times 10^{10}$ cm$^{-3}$. For a peak density around $10^{11}$ cm$^{-3}$ and considering an interaction volume determined by probe field propagation along the long axis with a cylindrical geometry of $\pi(0.2)^2 \times 14$ mm$^3$, we have $N \sim 1.8 \times 10^8$ with an optical density $\sim 400$.

- Numerical simulation of large N system is not possible, only up to several 10000.
- Local-field approx. too strong as an overestimation.
- Release of weak-field, local-field approximations, and ignorance of atom-atom correlations → Quantum Langevin eqs. / phase-space method simulations, a semiclassical method that includes quantum noises.

**Thank you for your attentions.**