



### Current PhD:

H. Busches  
C. Carr  
J. Keaveney  
D. Maxwell  
D. Parades

### Current PDRA:

D. Szwer

### Collaborators:

M. P. A. Jones (Durham)  
I. G. Hughes (Durham)  
R. M. Potvliege (Durham)  
K. J. Weatherill (Durham)  
D. Sarkisyan (Yerevan)  
M. Tanasittikosol (Bangkok)

### Sr Rydberg:

G. Lohead  
D. Boddy  
D. Sandler  
C. Vaillant



### Recent PhD:

R. Abel (2011)  
J. D. Pritchard (2011)

### Recent PDRA:

A. Gauguet (Toulouse)  
U. Krohn (Durham)  
A. Mohapatra (Bhubanesgar)



# In DIPOLES we deLIGHT

## Part I

Light field and one dipole

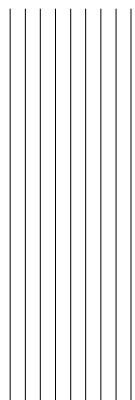
Dipole – dipole interactions

## Part II

Non-linear optics

Incident field

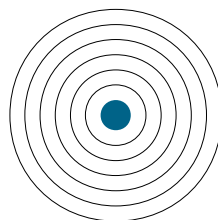
$$\mathcal{E}_i$$



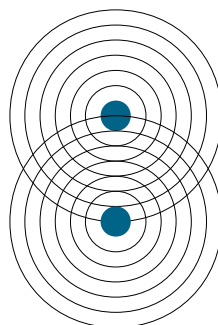
Induced dipole field

$$\mathcal{E}_d$$

One dipole

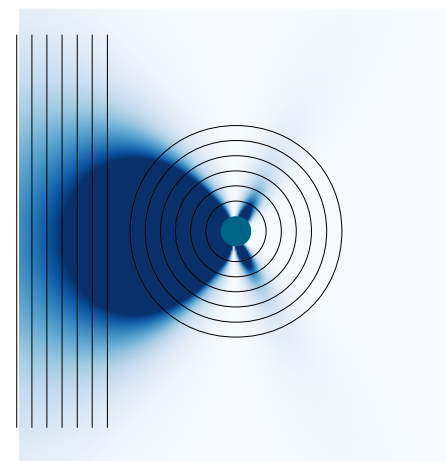


Two dipoles



Total field

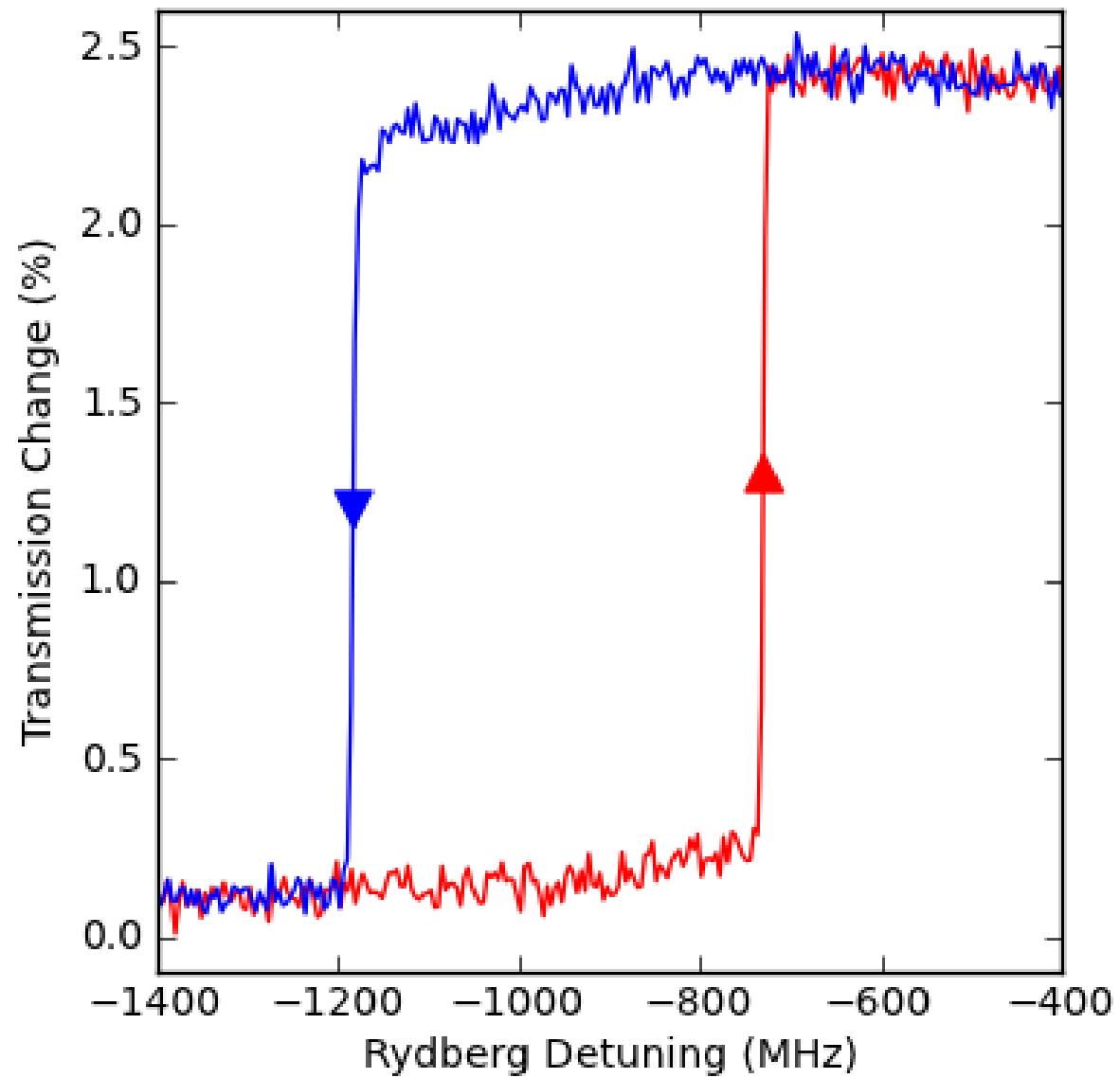
$$\mathcal{E}_t = \mathcal{E}_i + \mathcal{E}_d$$



Cooperative effects:

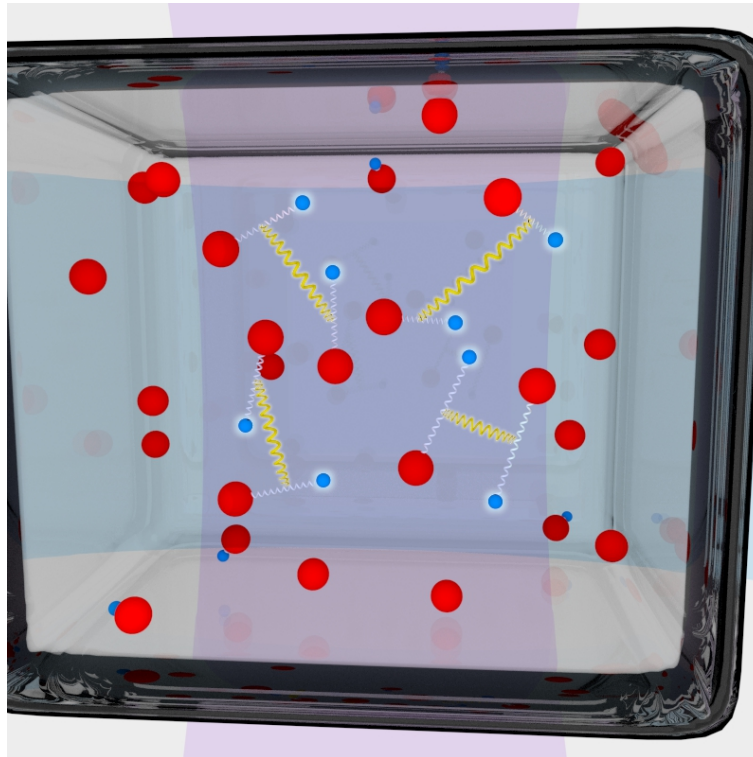
level shift/splitting, sub/superradiance

Strongly correlated quantum many body system!



Light induced dipole and dipole-dipole interactions, C. S. Adams  
Pisa Coherence School, September 17-20, 2012.

For intuition look to classical physics



Force  $\rightarrow \mathcal{E}$

Dielectric

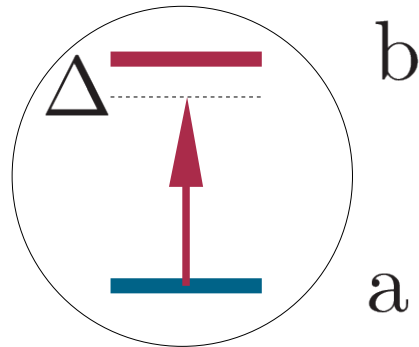
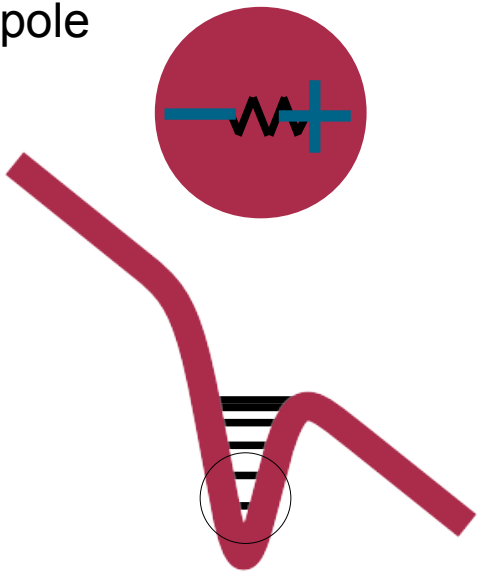


Displacement Force

$$P = N \langle d \rangle = \epsilon_0 \chi \mathcal{E}$$

Susceptibility: dimensionless

Dipole



$$\frac{\langle d \rangle}{4\pi\epsilon_0} = \alpha_p \mathcal{E} \quad \chi = 4\pi N \alpha_p$$

Medium      Single dipole

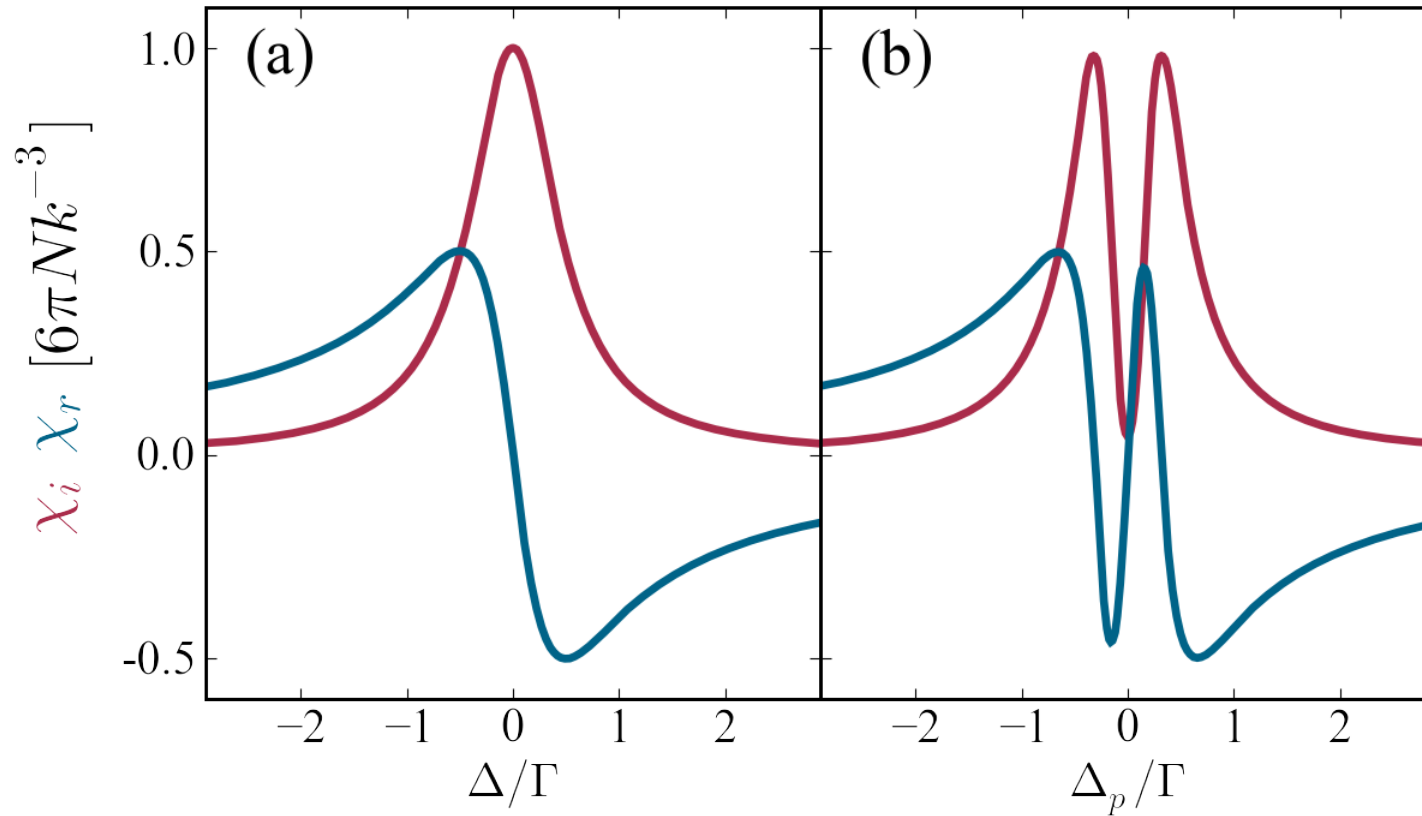
No dipole-dipole

$$\alpha_p = - \frac{1}{4\pi\epsilon_0} \frac{d_{ab}^2}{\hbar(\Delta + i\gamma_{ab})}$$

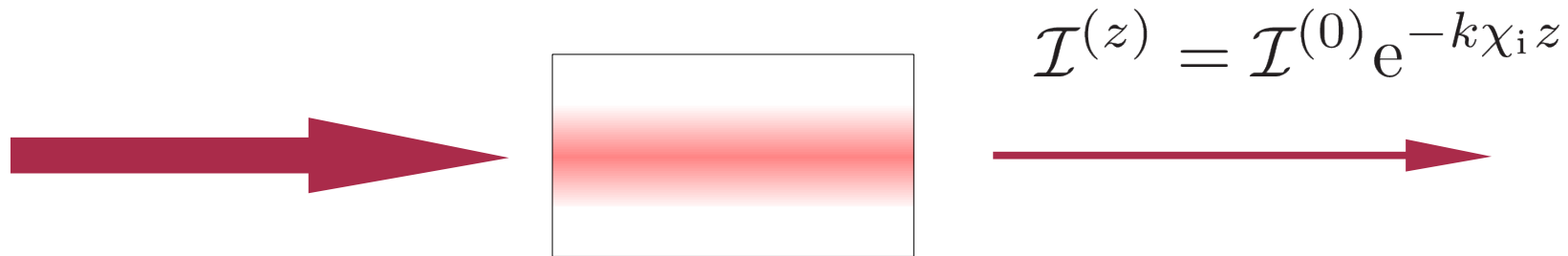
2-level

3-level

See Notes 5.4



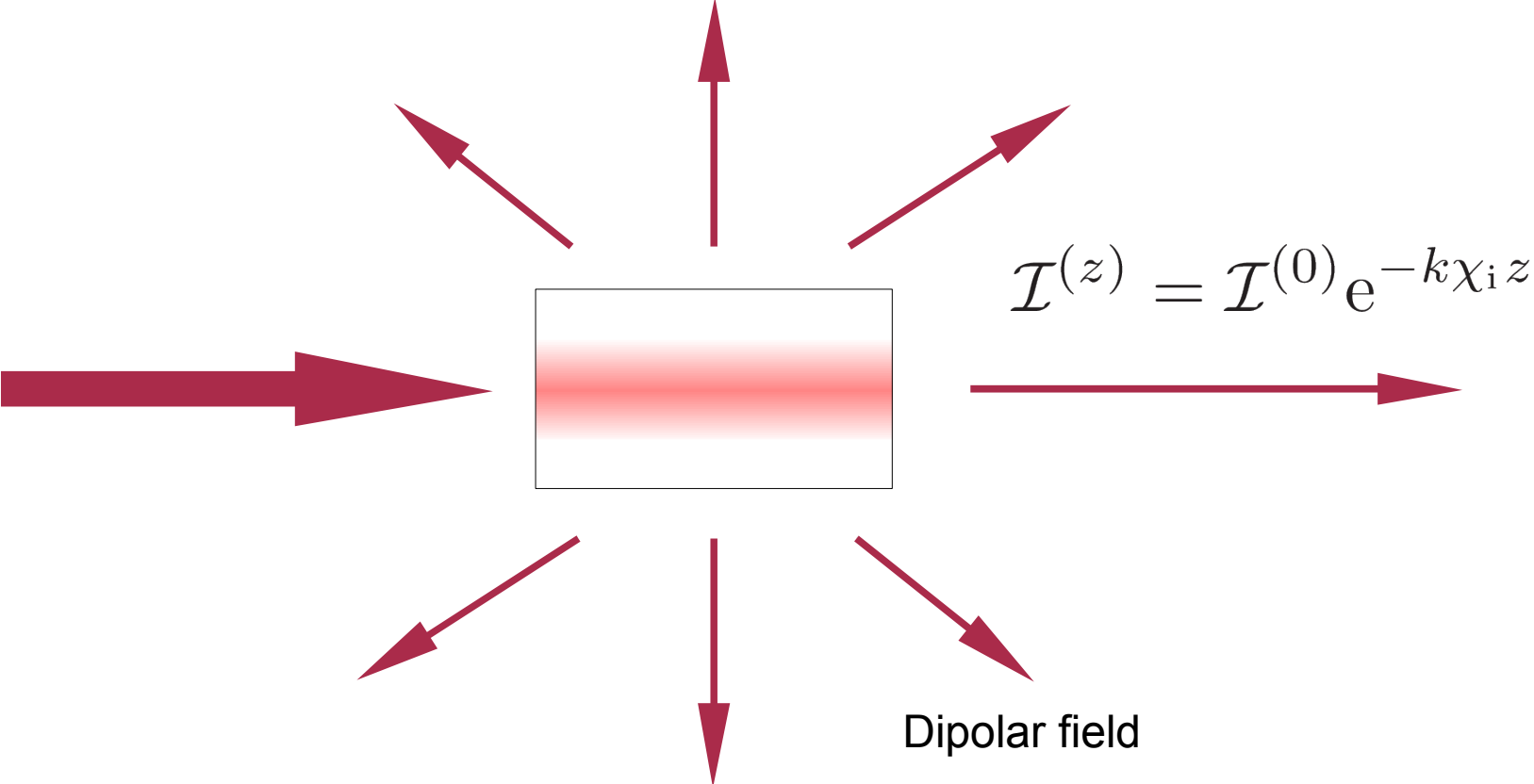




Aha! The light has been absorbed.

$$\alpha = k\chi_i$$

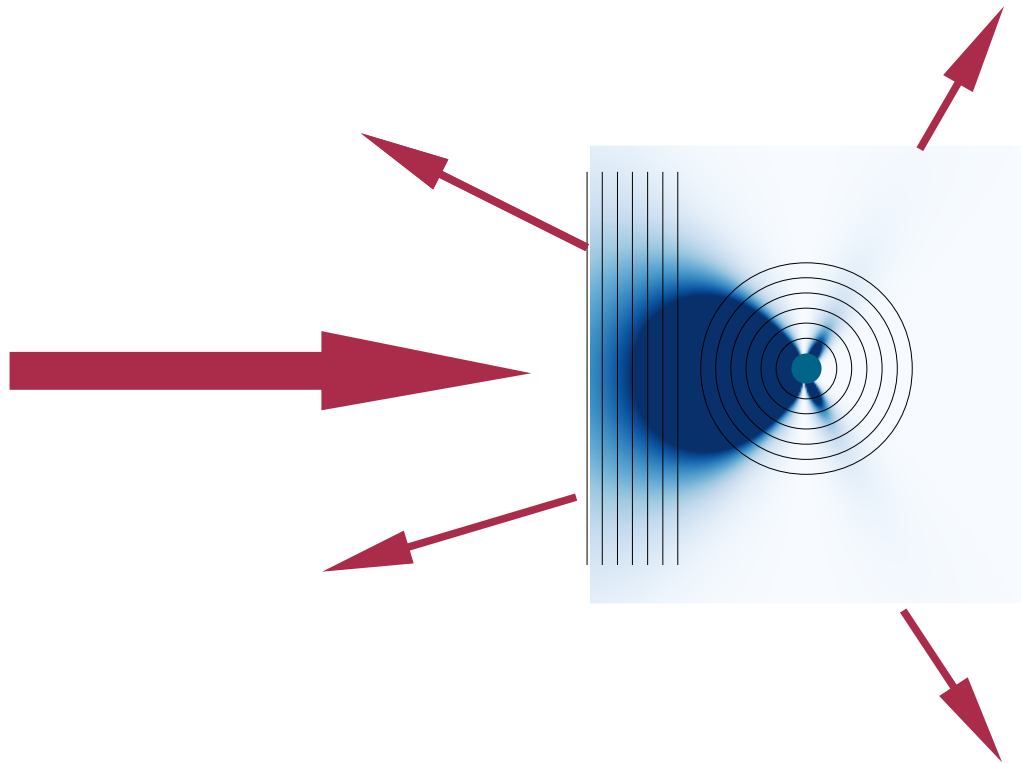
Where has the light gone?



No absorbed! Just scattered.

Is it possible to reduce the transmission to zero?

For a single dipole?

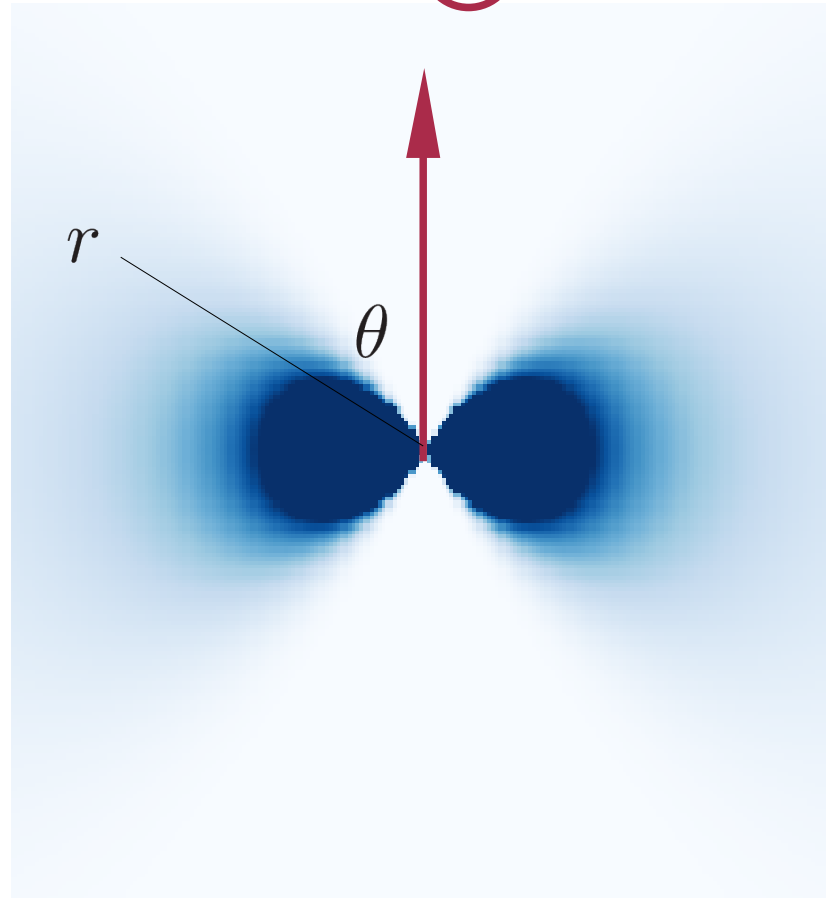


$$\mathcal{E}_t = \mathcal{E}_i + \mathcal{E}_d$$

Destruction interference  
in the  
forward scattering direction

## Electric field of a dipole

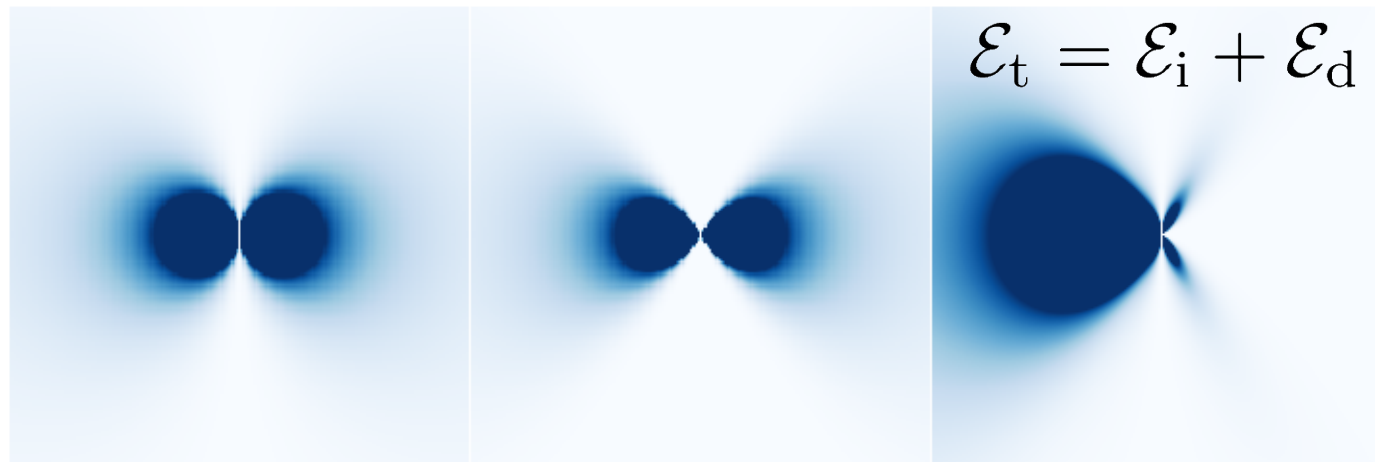
$$\mathcal{E}_z = \frac{d}{4\pi\epsilon_0} \left[ \left( \frac{1}{r^3} - \frac{ik}{r^2} \right) (3 \cos^2 \theta - 1) + \frac{k^2}{r} \sin^2 \theta \right] e^{i(kr - \omega t)}$$



## Focussed Gaussian

$$\mathcal{E}_i^{(z)} = \left(-i\right) \frac{zR}{z} \mathcal{E}_0 e^{ikz} e^{ik\rho^2/2z} e^{-\rho^2/w_0^2}$$

A diffraction light field acquires a phase of  $\pi/2$

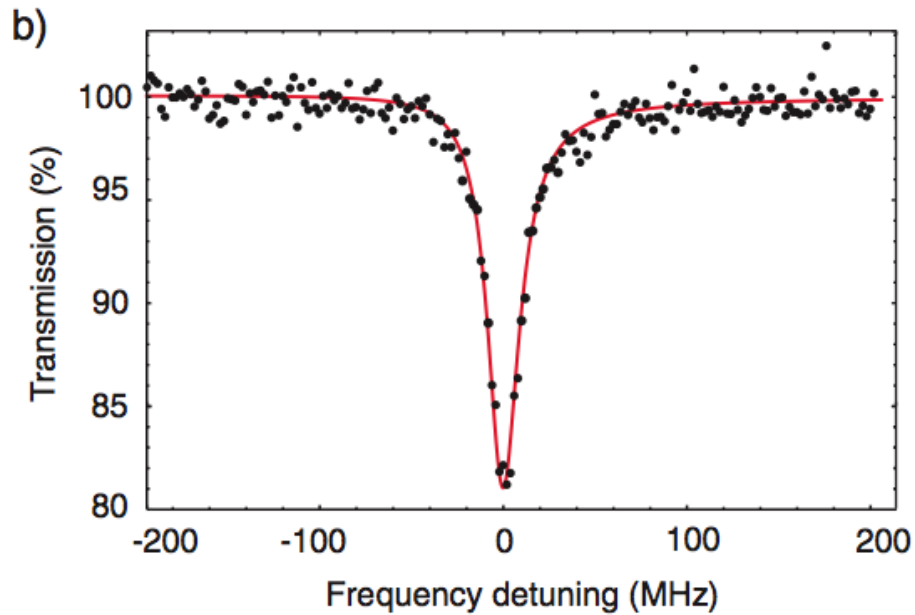
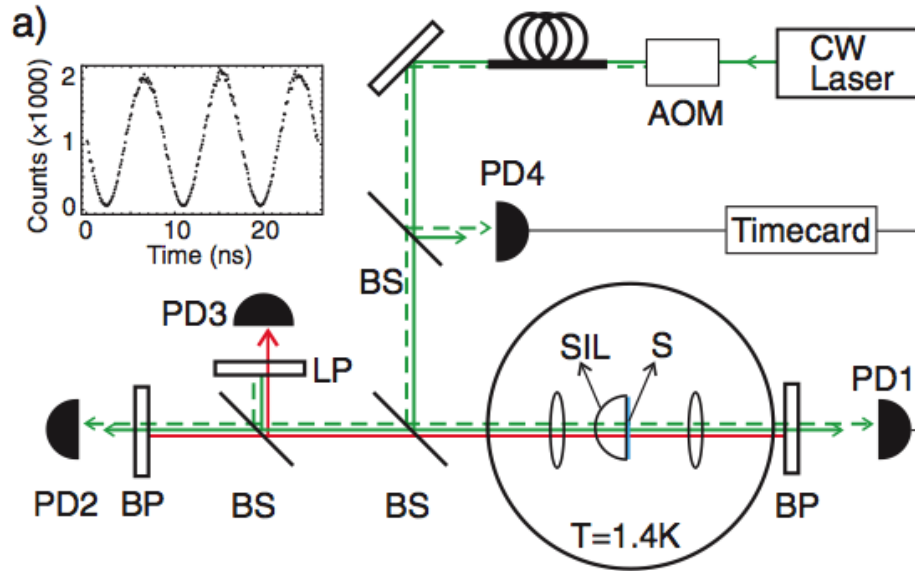


Dipole field

$$\mathcal{E}_d = \left(-i\right) \frac{3}{2} \frac{1}{kr} \mathcal{E}_0 \sin^2 \theta e^{i(kr - \omega t)}$$

On resonance a driven oscillator lags the driving field by  $\pi/2$

State of the art



Pototschnig *et al.*  
 Phys. Rev. Lett. **107**, 063001 (2012).

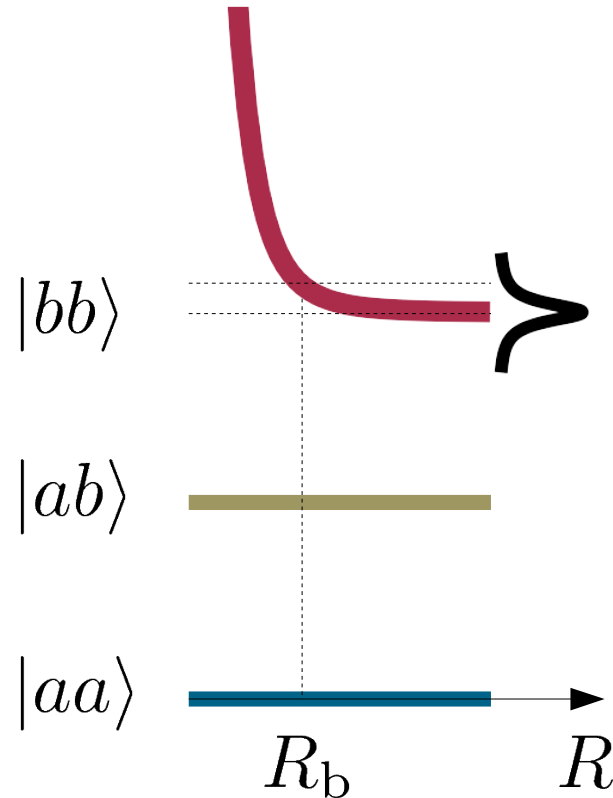
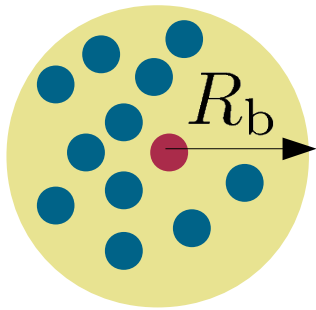
See also work by:

Kurtsiefer (Singapore)  
 Leuch (Erlangen)

An ensemble of  $N$  dipoles that only support a single excitation.

An ensemble of  $N$  dipoles that only support a single excitation.

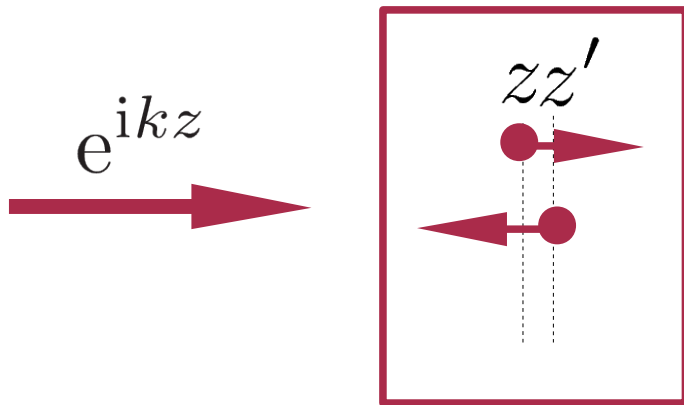
A Rydberg superatom



$$|\psi\rangle = \frac{1}{\sqrt{N}} \sum_{j=1}^N |a_1 \dots b_j \dots a_N\rangle$$

$$\sigma = N \frac{3\lambda^2}{2\pi}$$



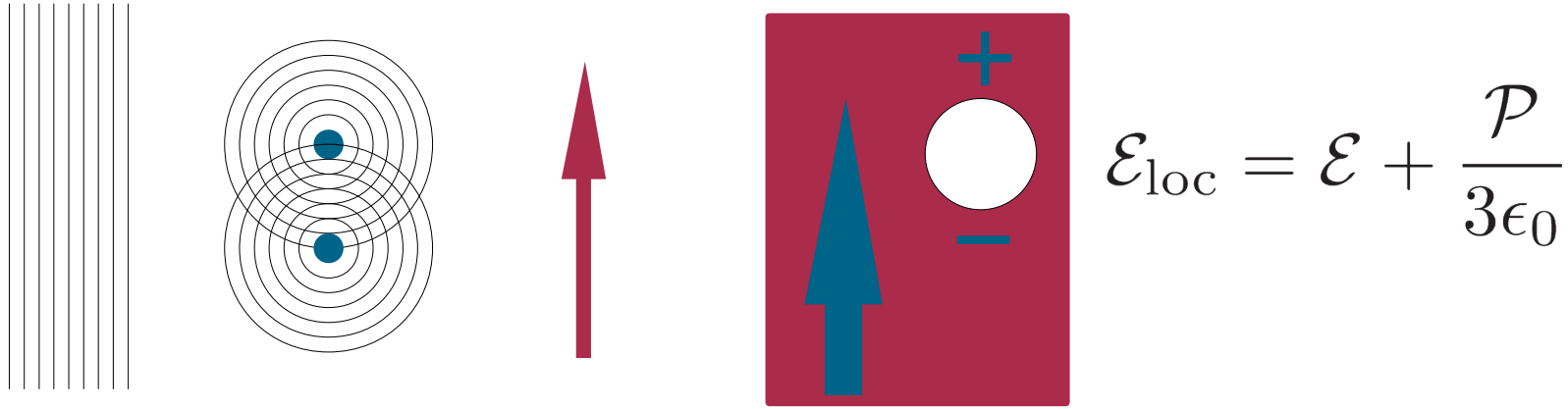


$$\mathcal{E}'_0 e^{ik'z} = \mathcal{E}_0 e^{ikz} + i \frac{k\chi}{2} e^{ikz} \int_z^\infty dz' \mathcal{E}'_0 e^{i(k'-k)z'} + i \frac{k\chi}{2} e^{-ikz} \int_0^z dz' \mathcal{E}'_0 e^{i(k'+k)z'}$$

Incident field
Forward dipolar field
Backward dipolar field

$$\mathcal{E}_t^{(z)} = \mathcal{E}_0 e^{ikz} - \mathcal{E}_0 e^{ikz} + \frac{2}{n+1} \mathcal{E}_0 e^{inkz} \quad n = \sqrt{1+\chi}$$

The superposition of the incident field and the dipolar field inside the medium is plane wave with reduced amplitude and modified phase velocity.



$$\mathcal{P} = \epsilon_0 \chi \mathcal{E}$$

$$\mathcal{P} = 4\pi\epsilon_0 N \alpha_p \mathcal{E}_{\text{loc}}$$

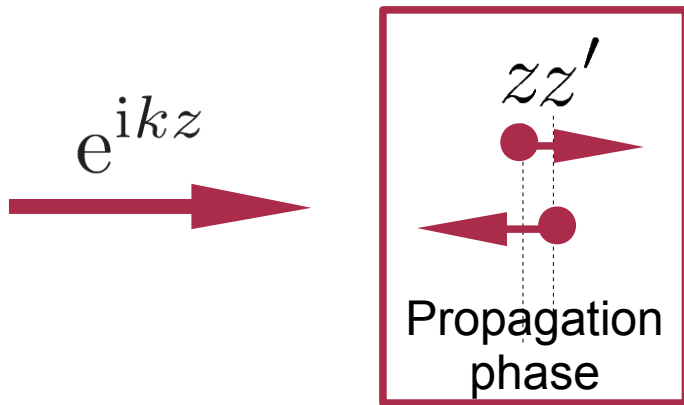
$$\chi = \frac{4\pi N \alpha_p}{1 - \frac{4}{3}\pi N \alpha_p}$$

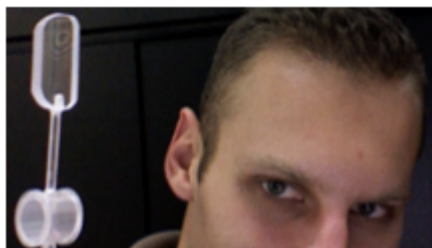
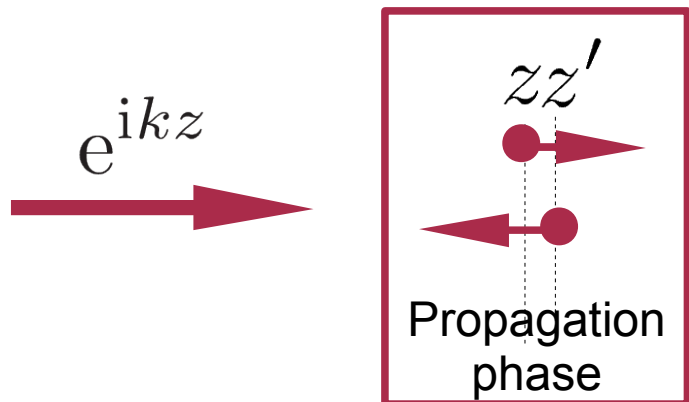
Lorentz-Lorenz law

Also Clausius Mossotti

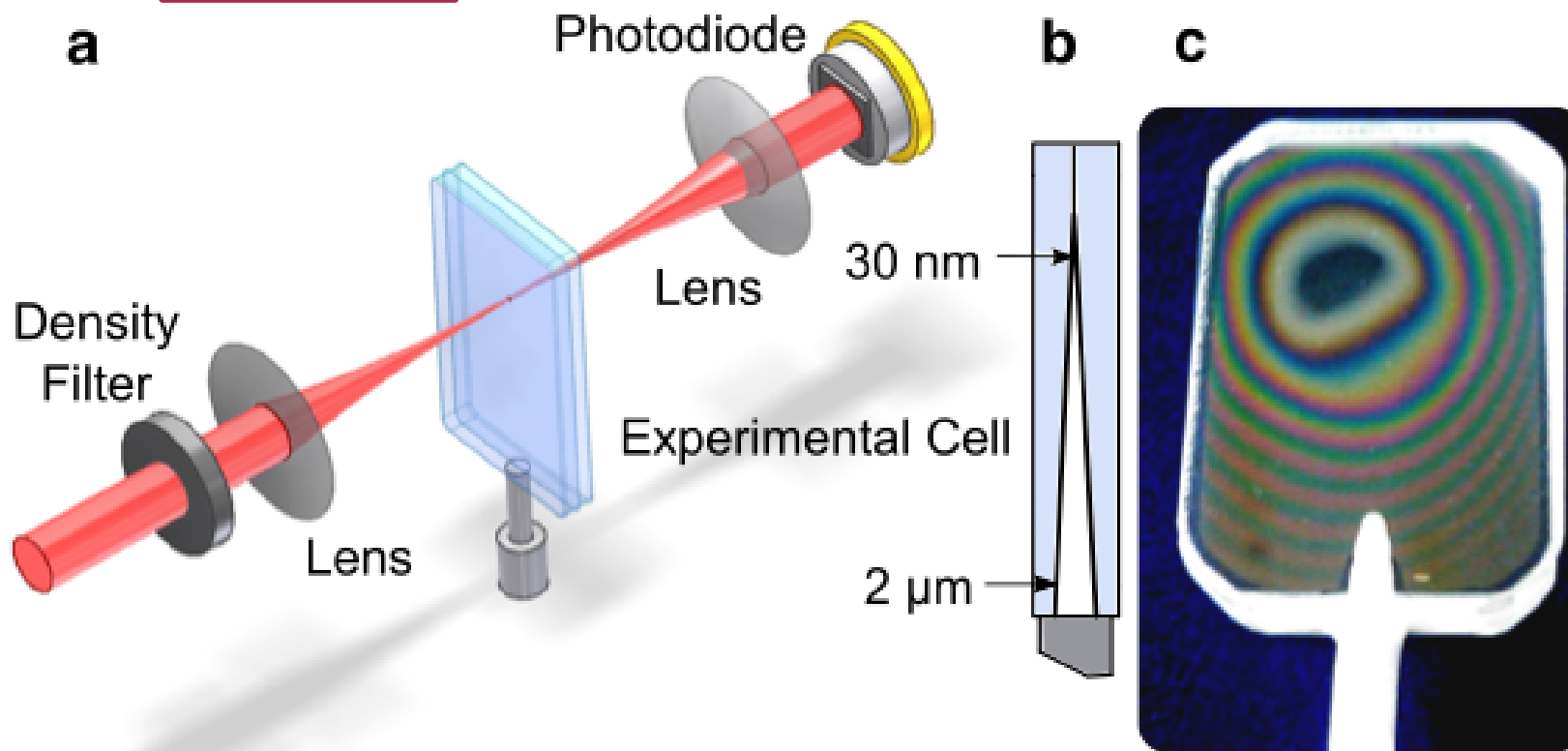
$$\chi = - \frac{N d^2 / \epsilon_0 \hbar}{\Delta + i\gamma_{ab} + \underbrace{N d^2 / 3\epsilon_0 \hbar}}_{\text{Lorentz shift}}$$

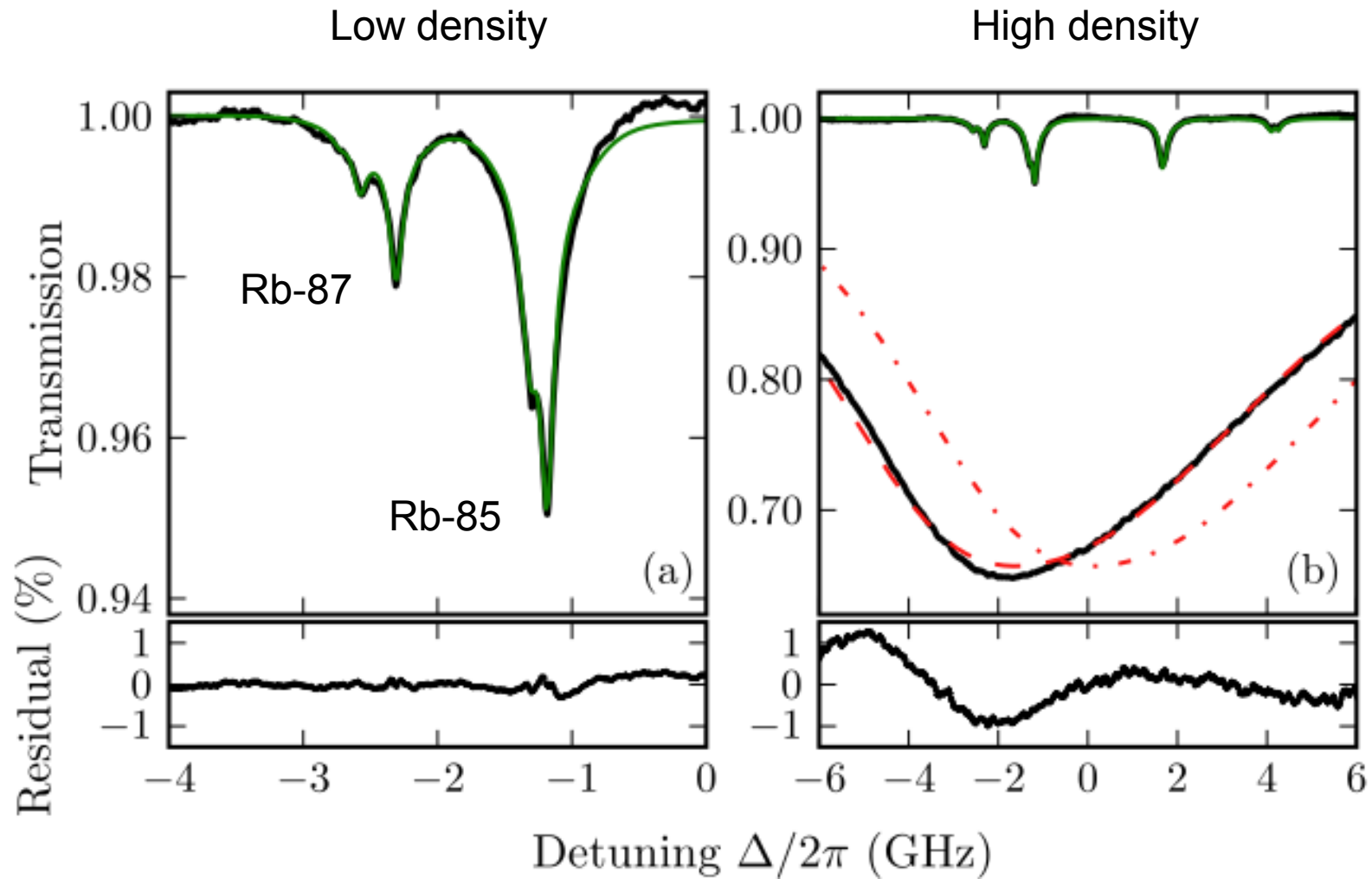
Lorentz shift



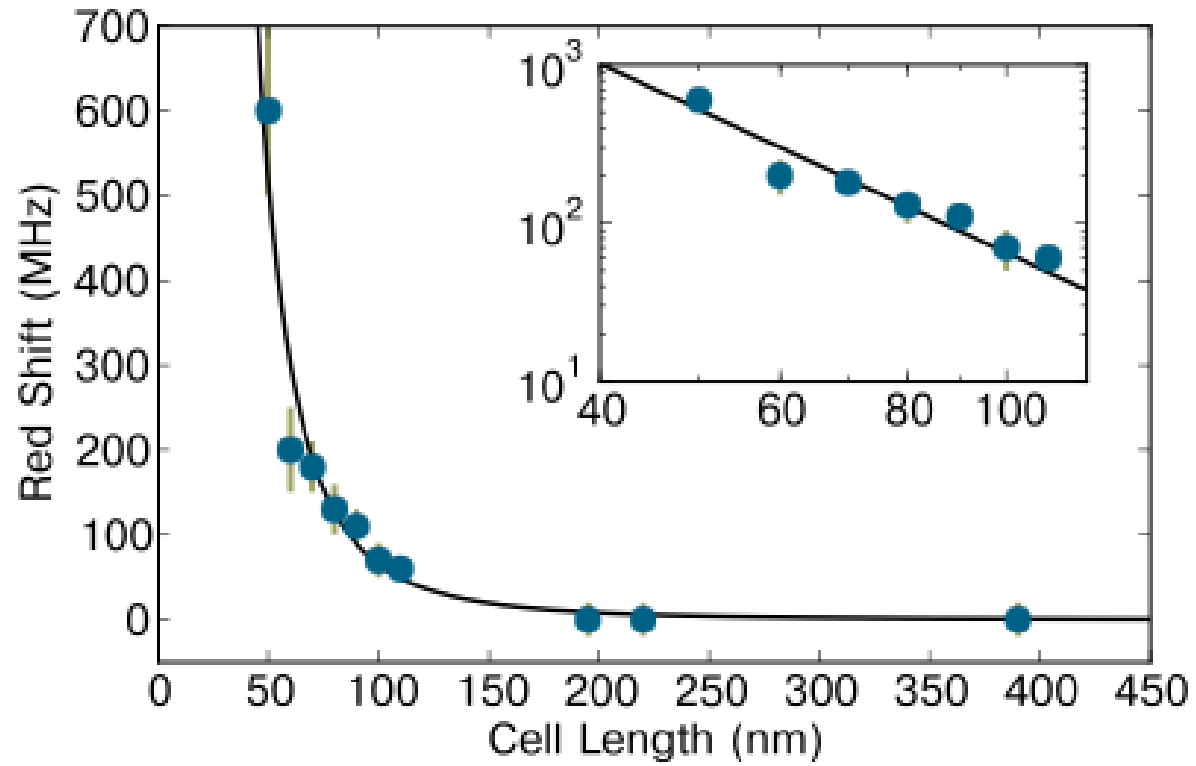


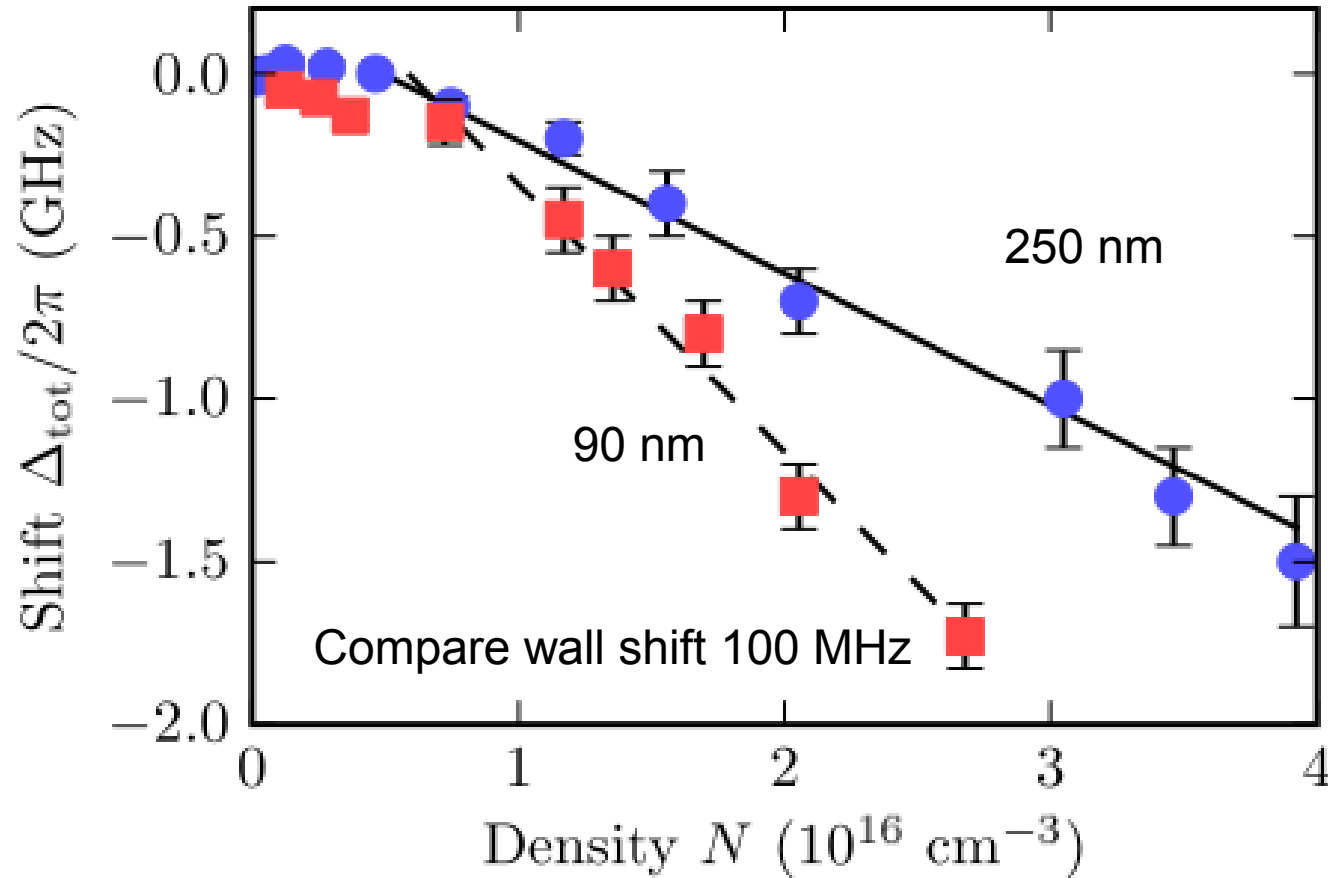
David Sarkisyan, Armen Sargsyan,  
Armenian Academy of Sciences



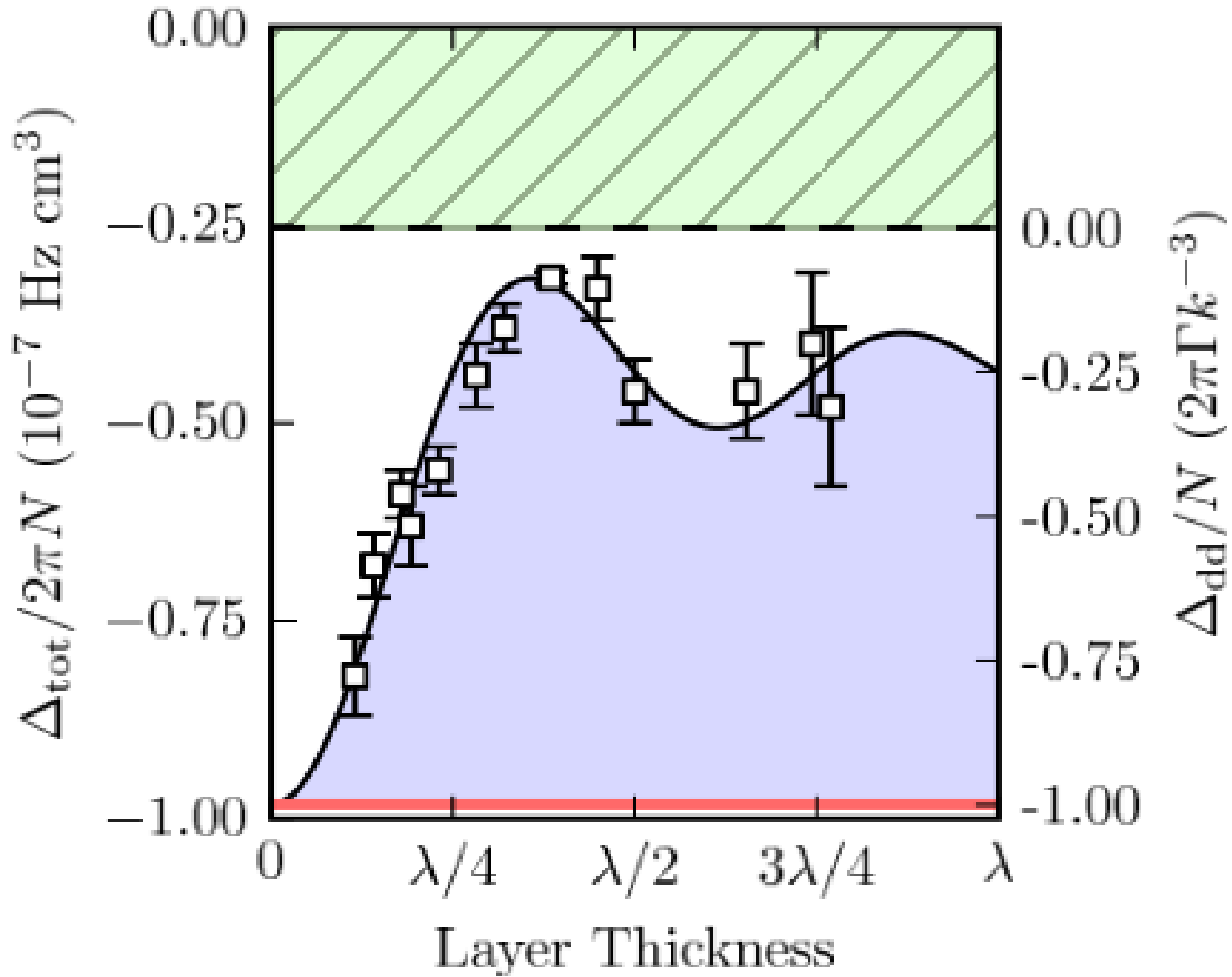


J. Keaveney *et al.*, arXiv:1201.5251





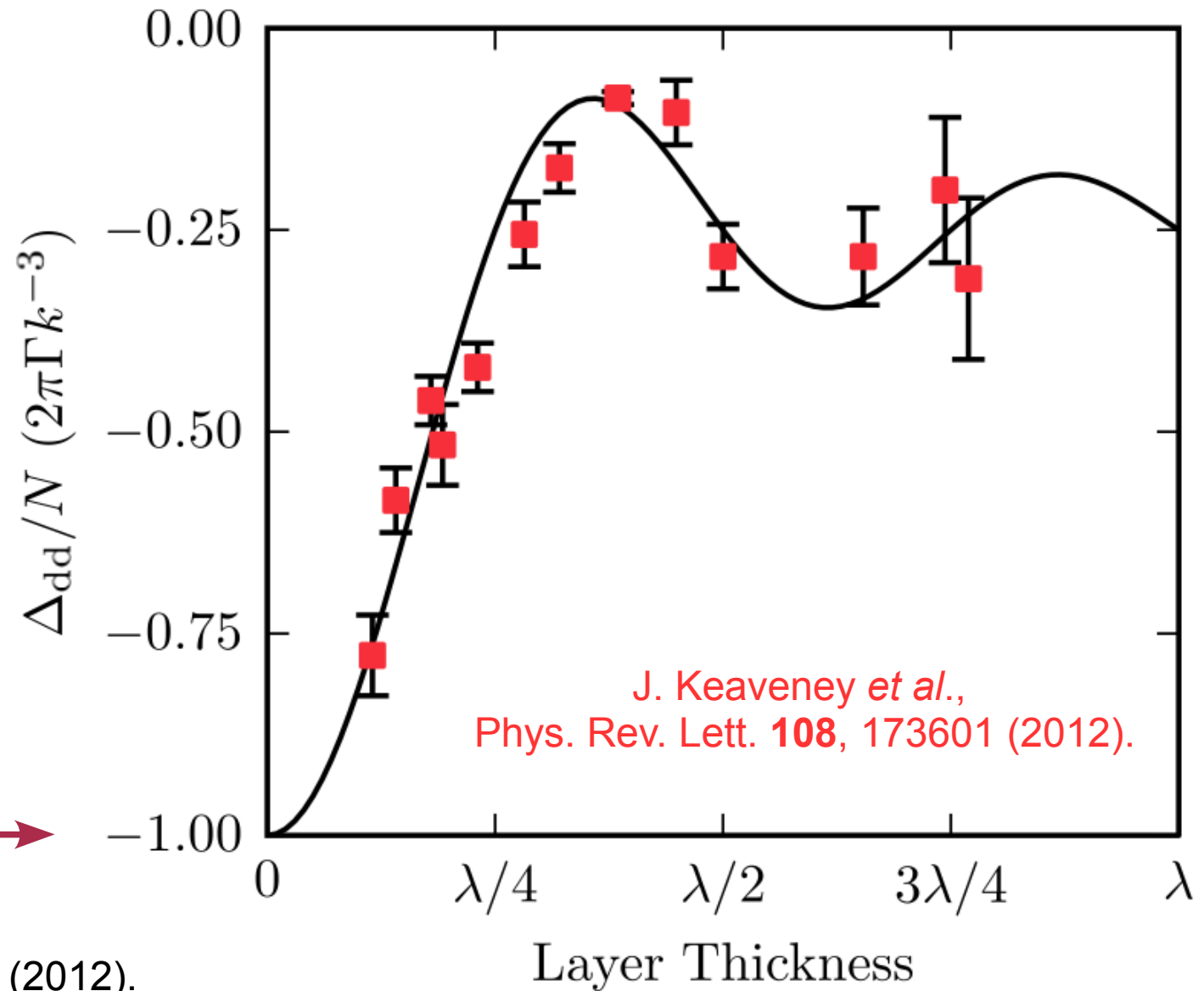
Note the high density!



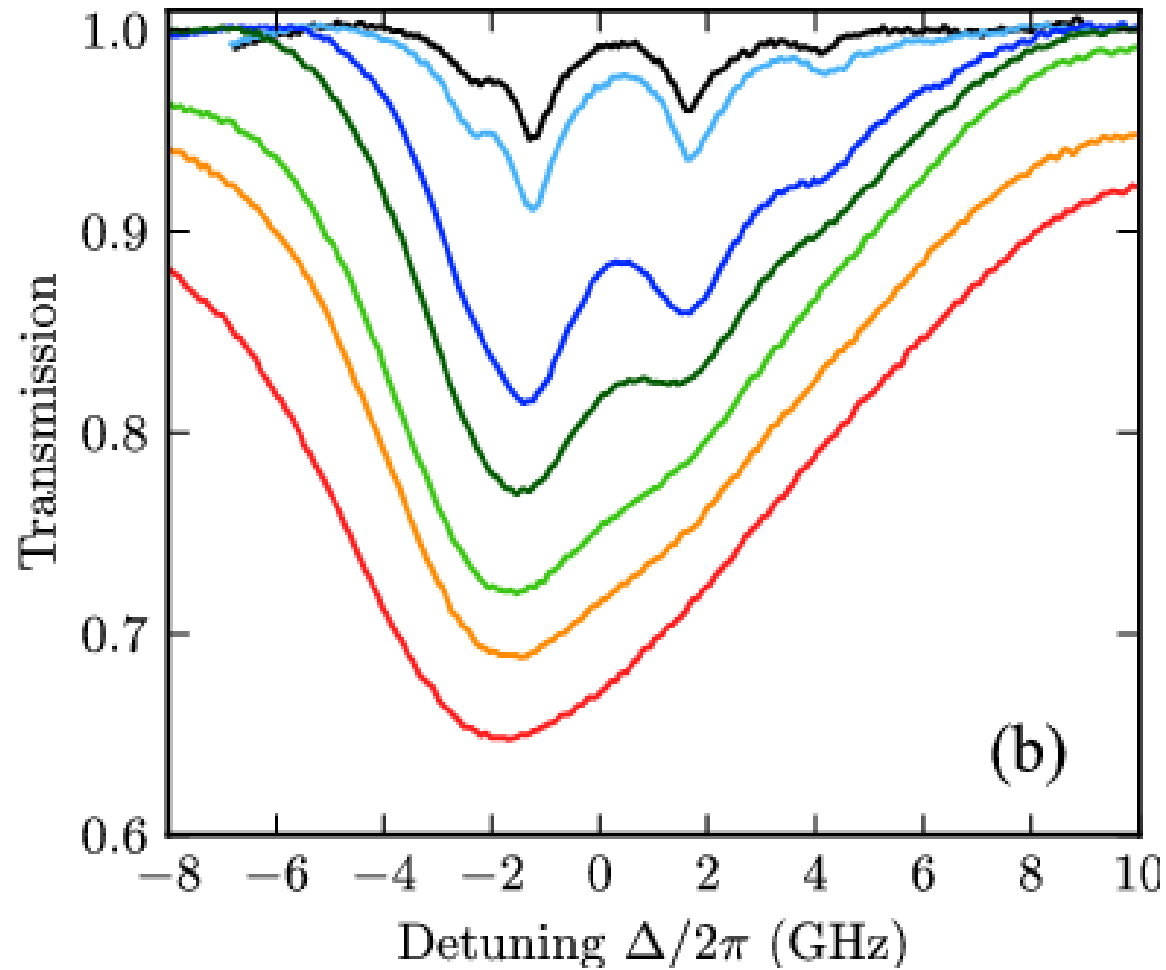


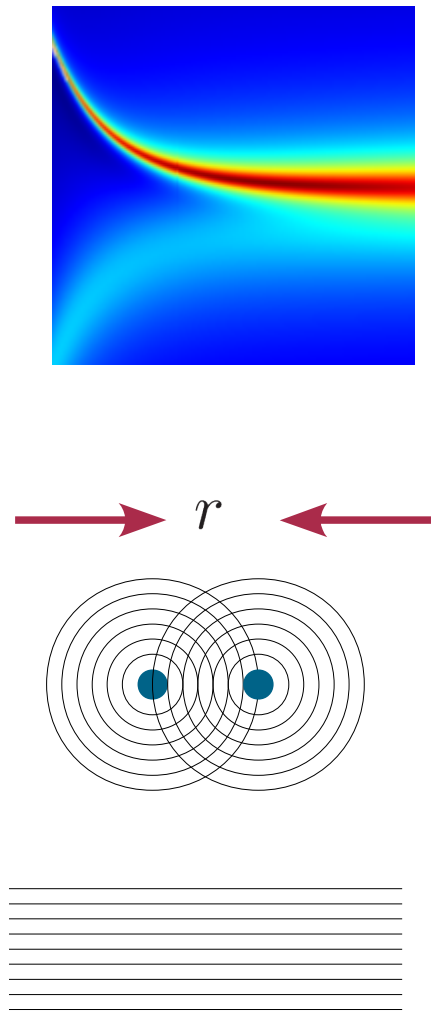
R. Friedberg *et al.*  
 Phys. Rep. **7**, 101 (1973).

Lorentz shift →

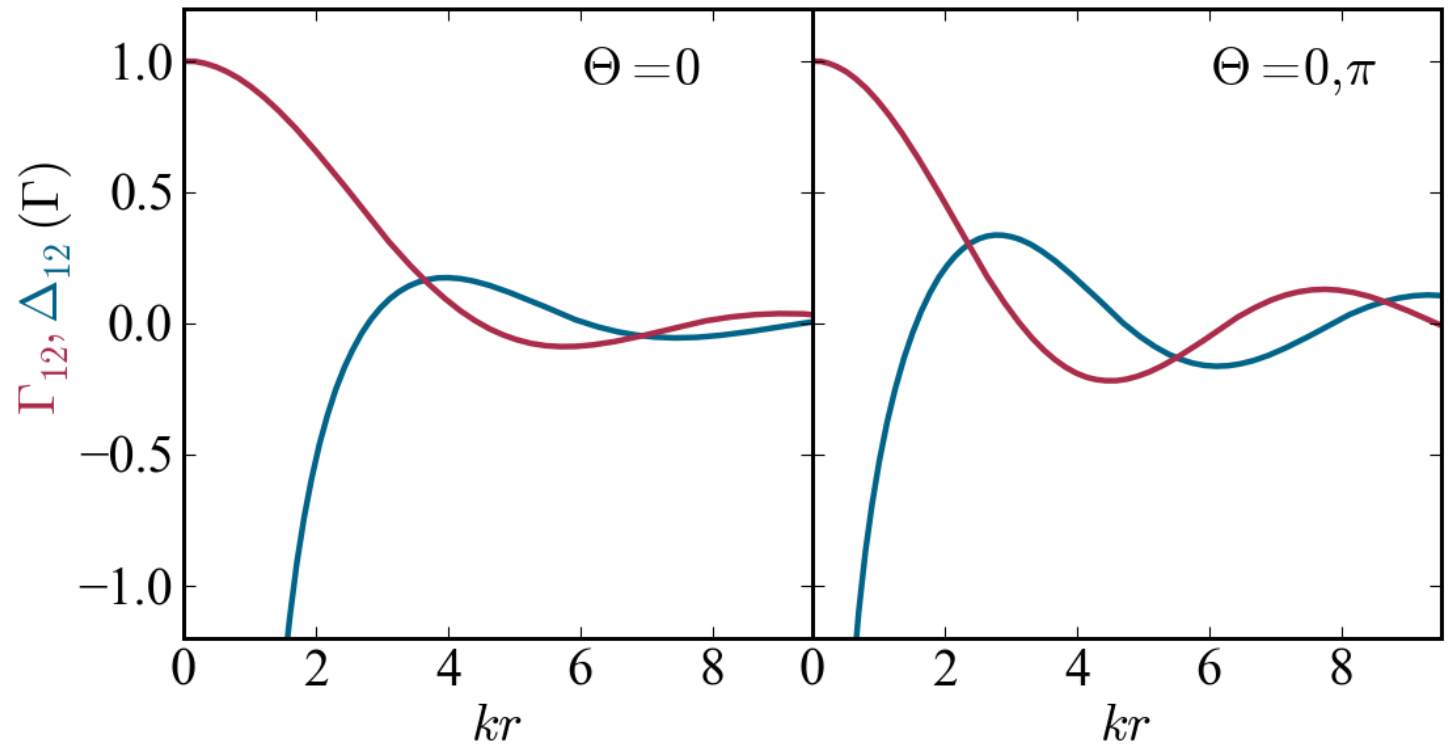


R. Rohlsberger., Physics. **5**, 46 (2012).





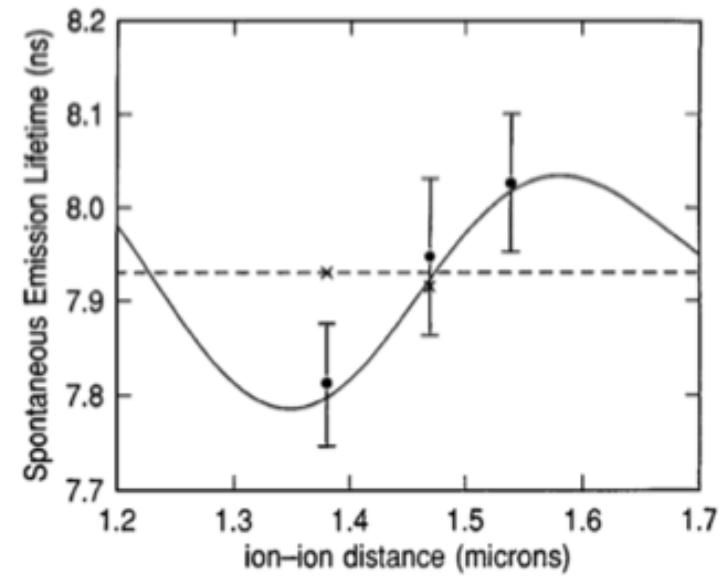
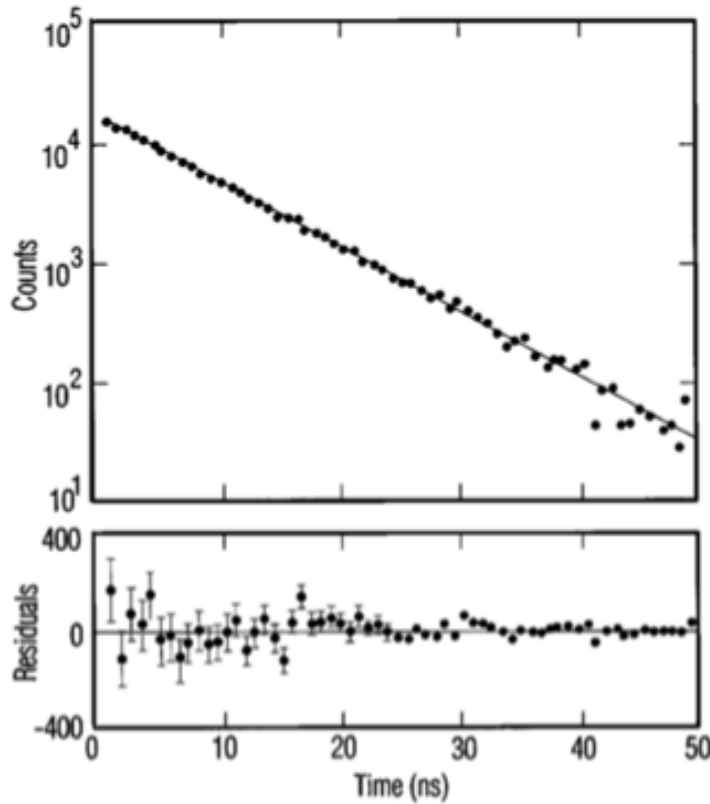
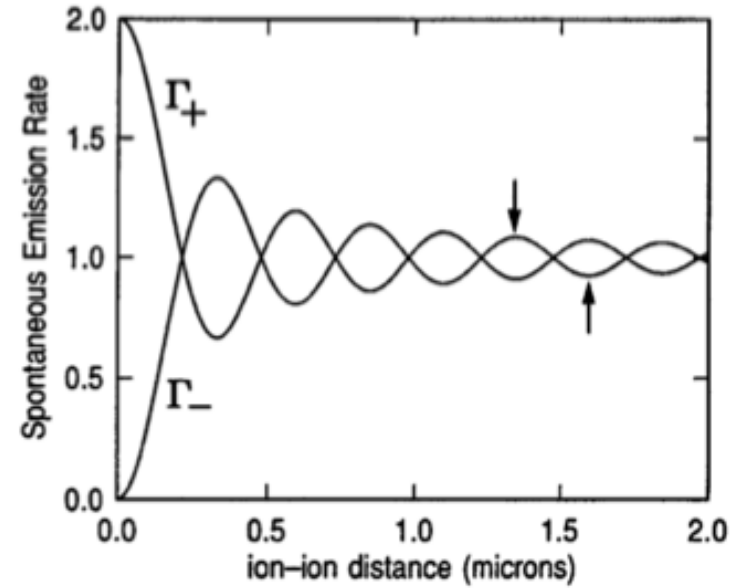
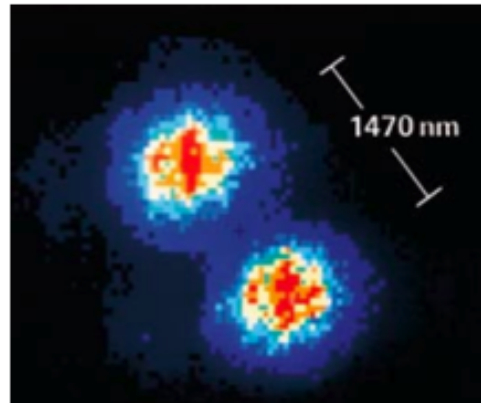
$$|\psi\rangle = \frac{1}{\sqrt{2}}(|ab\rangle \pm |ba\rangle)$$

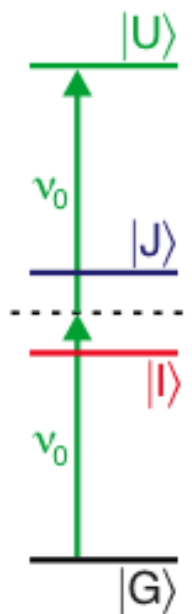
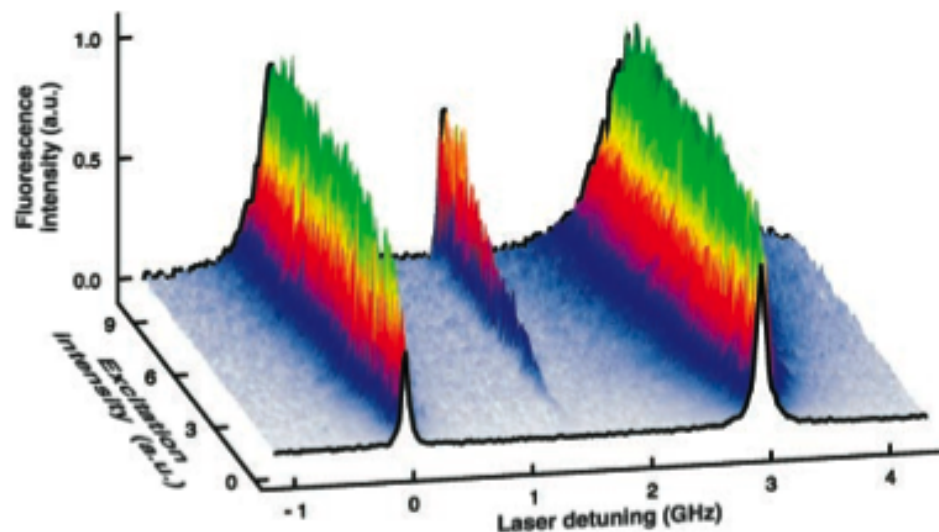
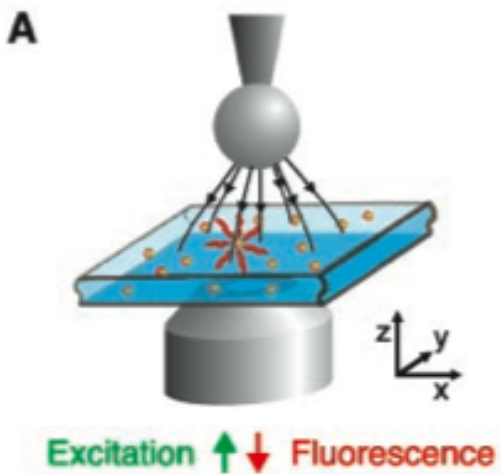


Near field  $kr < 1$

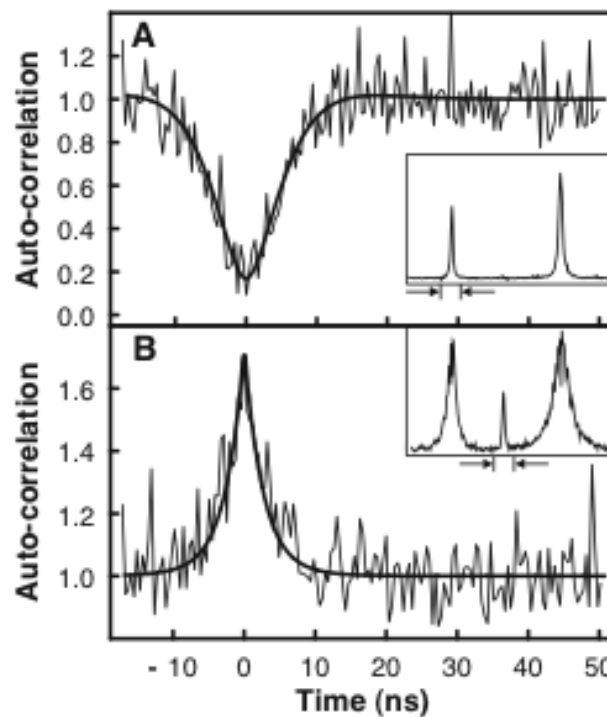
Optical dipoles  $r < 100$  nm

Rydberg dipoles  $r < 1$  mm





Coupled system

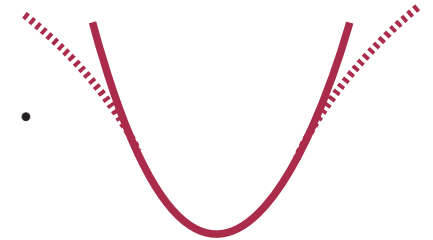


1. Always think in terms of the superposition of the incident field and the induced dipole field.
2. There is no absorption!  
Only destructive interference in the forward scattering direction.
3. The extinction problem.  
Imperfect coupling between a photon and a single dipole.

The interaction depends on the incident light intensity.

Linear optics  $P = \epsilon_0 \chi \mathcal{E}$  Displacement is linearly proportional to the force

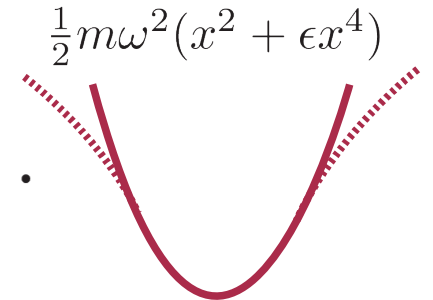
Non-linear optics  $P = \epsilon_0 \chi^{(1)} \mathcal{E} + \epsilon_0 \chi^{(3)} \mathcal{E}^3 + \dots$



The interaction depends on the incident light intensity.

Linear optics  $P = \epsilon_0 \chi \mathcal{E}$  Displacement is linearly proportional to the force

Non-linear optics  $P = \epsilon_0 \chi^{(1)} \mathcal{E} + \epsilon_0 \chi^{(3)} \mathcal{E}^3 + \dots$

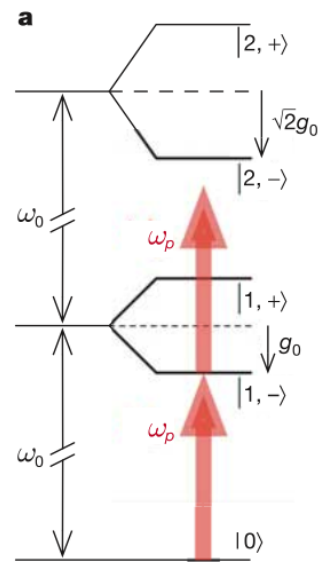


Quantum: single photon non-linearity anharmonic level spacing.

1. ac Stark shift

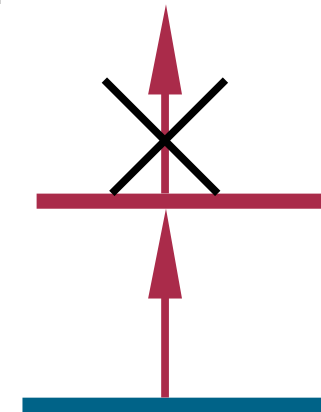
Cavity QED

Birnbaum *et al.*  
Nature **436**, 101 (2005).



2. Saturation

Single dipole

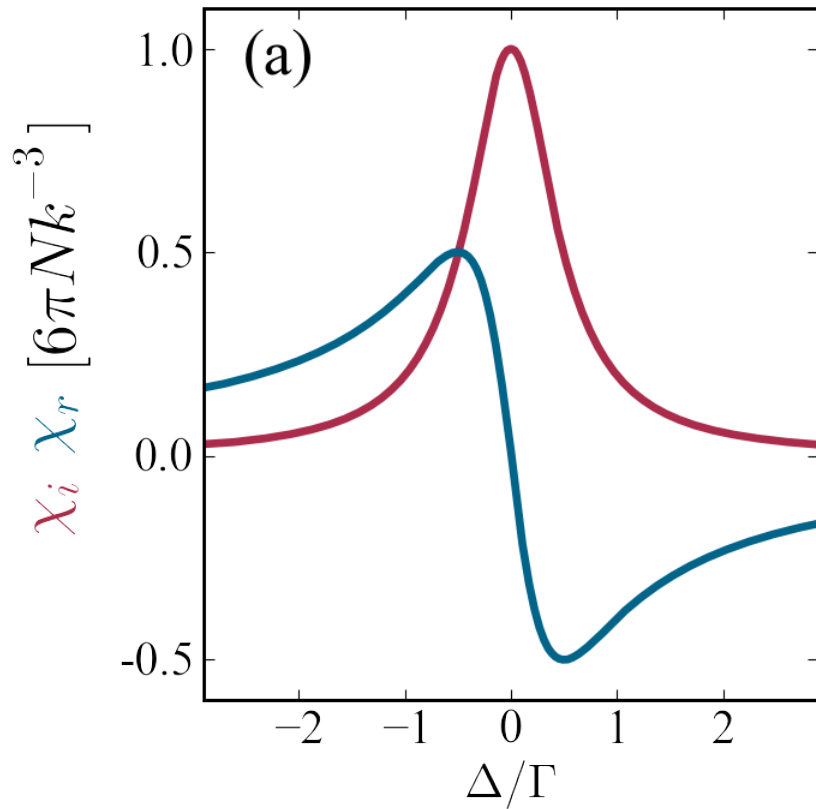


Single atom  
Single molecule

The extinction problem

Rydberg superatom





$$\chi_r = \chi_r^{(1)} + \chi_r^{(3)} \mathcal{E}^2$$

$$\chi_r = \chi_r^{(1)} + \frac{\partial \chi_r}{\partial \omega} \Delta\omega$$

$$n_g = 1 + \frac{\omega}{2} \frac{\partial \chi_r}{\partial \omega}$$

ac Stark shift

$$\Delta\omega = -\frac{1}{2} \frac{\alpha_p \mathcal{E}^2}{\hbar}$$

$$\chi^{(3)} = \frac{(n_g - 1)\alpha_p}{\hbar\omega}$$

Far off resonance

$$\chi_r^{(3)} = \chi_r^{(1)} \frac{d_{ab}^2}{(\hbar\omega_0)^2}$$

$$\mathcal{E}_{at} = \hbar\omega_0/d_{ab}$$

$$\chi_r^{(3)} = \frac{\chi_r^{(1)}}{\mathcal{E}_{at}^2}$$

Binding field

$$\mathcal{E}_{at} \sim \frac{1}{2}e/4\pi\epsilon_0 a_0^2 \sim 5 \times 10^{11} \text{ Vm}^{-1}$$

Non-linear term

$$\chi_r^{(3)} \mathcal{E}^2 = \frac{\chi_r^{(1)}}{\mathcal{E}_{at}^2} \mathcal{E}^2$$

Ratio of incident field  
to binding field squared

$$\left| \chi_r^{(3)} \right| \leq 10^{-23} \text{V}^{-2} \text{m}^2$$

Water

$$\chi_r^{(3)} = 2.5 \times 10^{-22} \text{V}^{-2} \text{m}^2$$

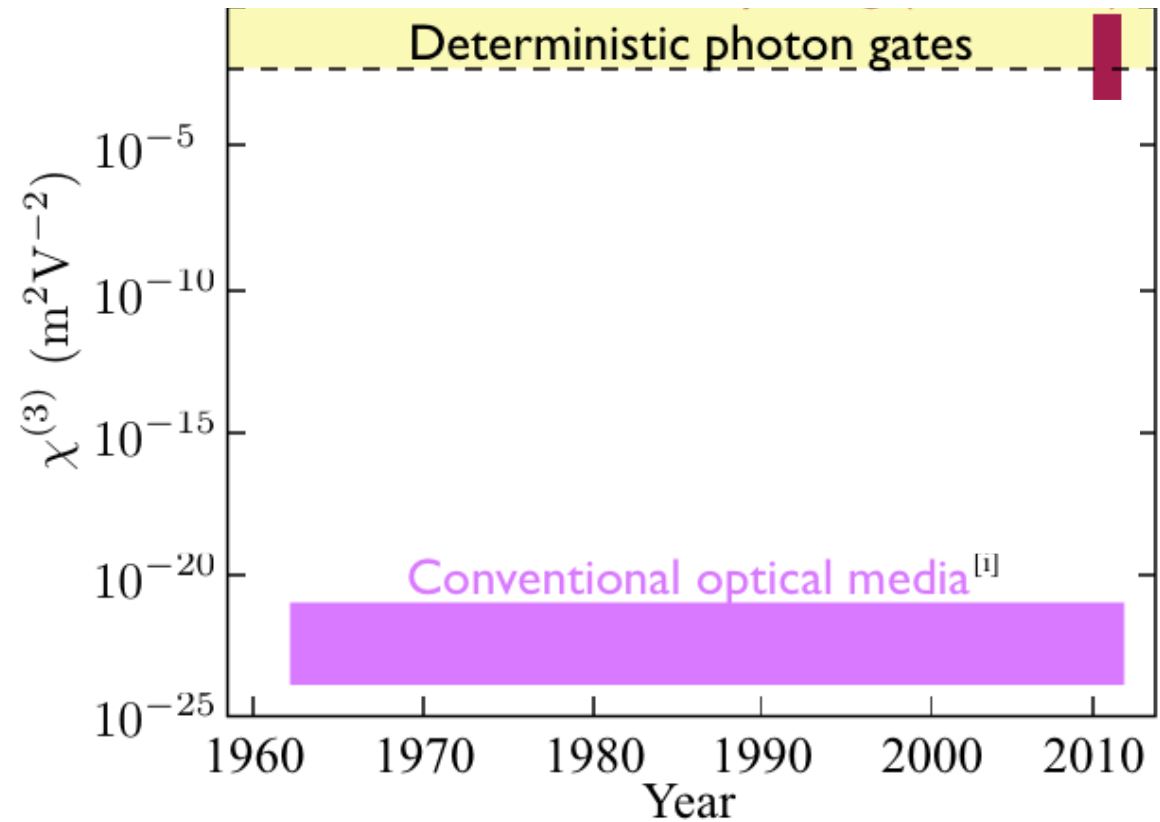
E field of a photon  $\hbar\omega_0 = \frac{1}{2}\epsilon_0\mathcal{E}^2\pi w_0^2\ell$

Length 1 MHz bandwidth 1 micron in a fibre

$$\mathcal{E} \sim 3 \times 10^1 \text{ Vm}^{-1}$$

$$\mathcal{E}_{\text{at}} \sim 5 \times 10^{11} \text{ Vm}^{-1}$$

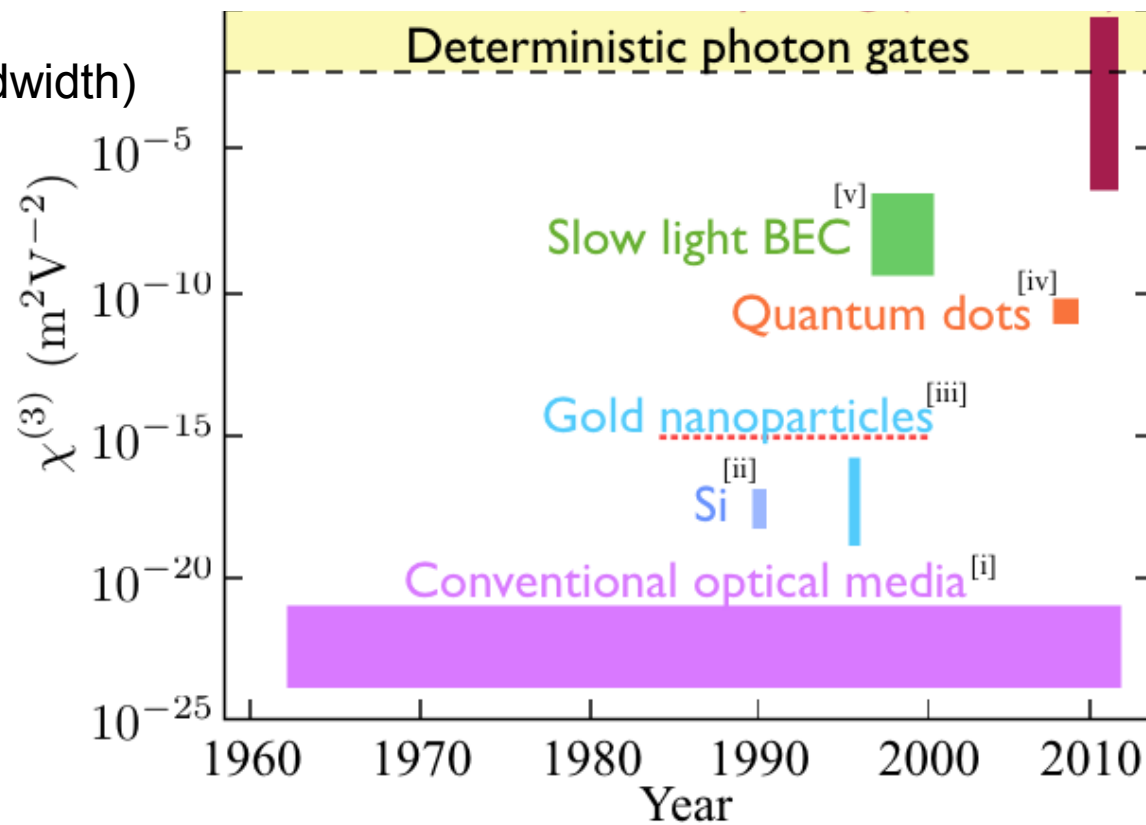
$$\chi_r^{(3)}\mathcal{E}^2 = \frac{\chi_r^{(1)}}{\mathcal{E}_{\text{at}}^2}\mathcal{E}^2$$

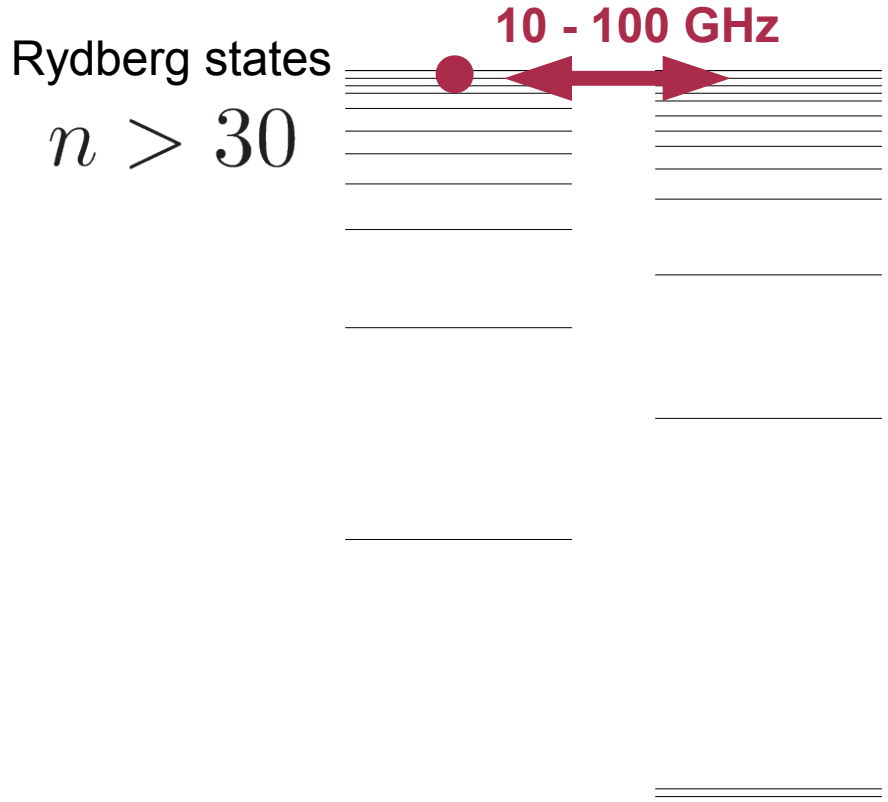


$$\chi^{(3)} = \frac{(n_g - 1)\alpha_p}{\hbar\omega}$$

Resonance (enhancement but reduced bandwidth)

$$\chi^{(3)} \sim \chi^{(1)} \frac{\Gamma}{\Delta} \left( \frac{d\mathcal{E}_{ph}}{\hbar\Omega_c} \right)^2$$





Rydberg states

Size  $\langle r \rangle \sim n^2 a_0$

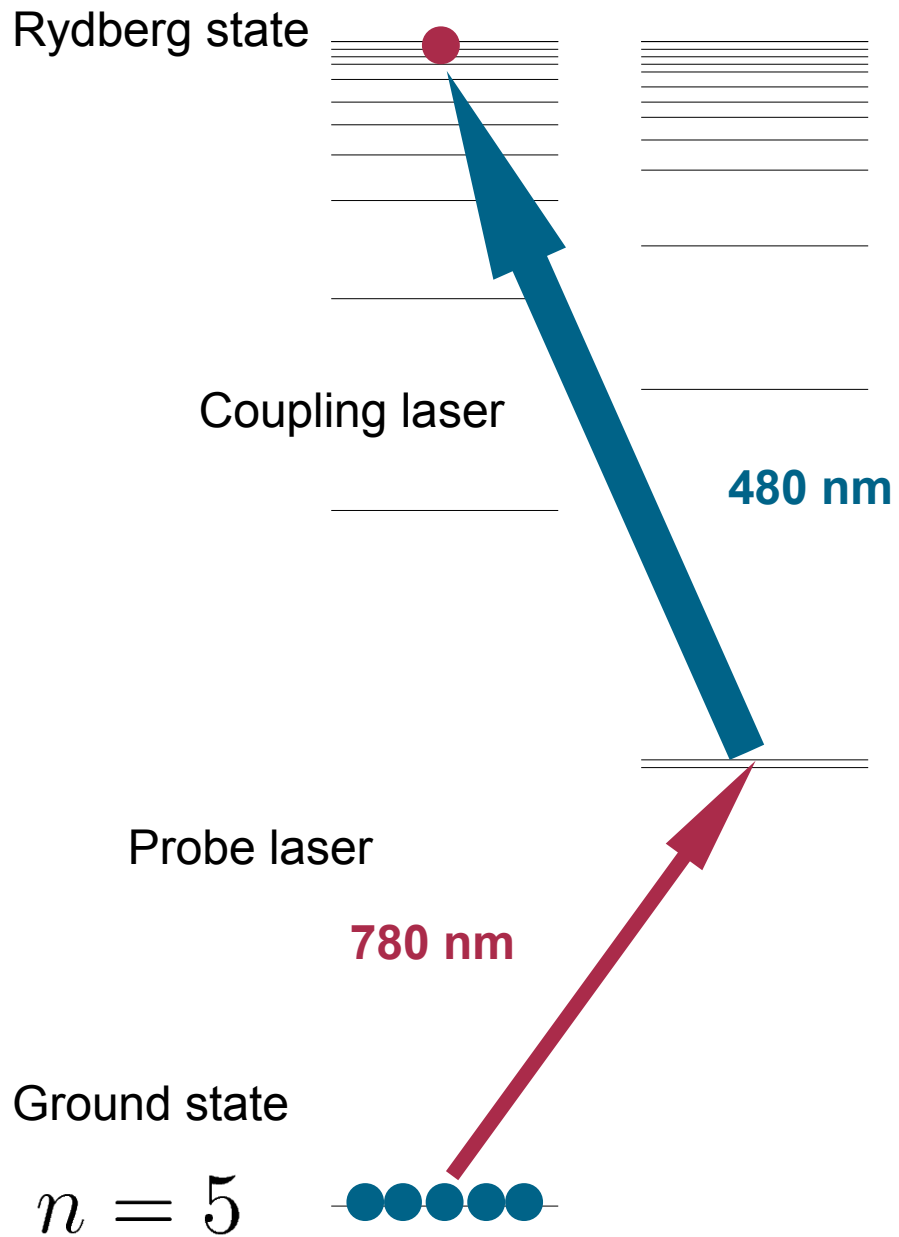
Dipole moment  $\langle d \rangle \sim n^2 e a_0$

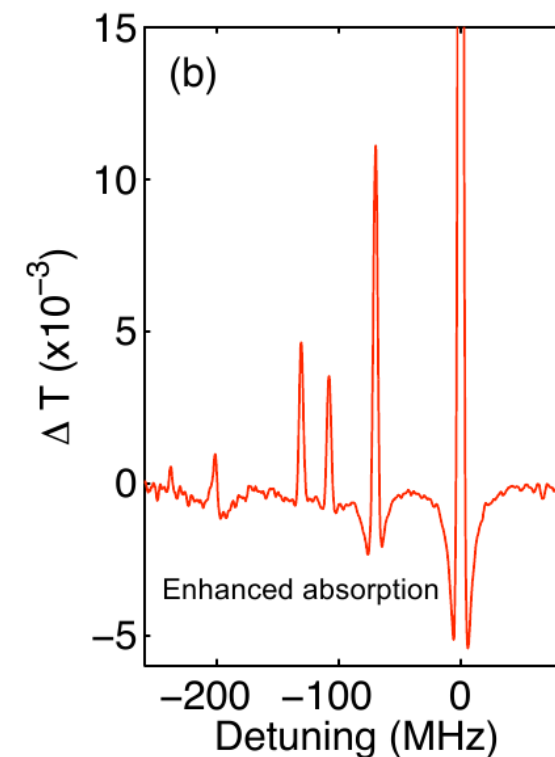
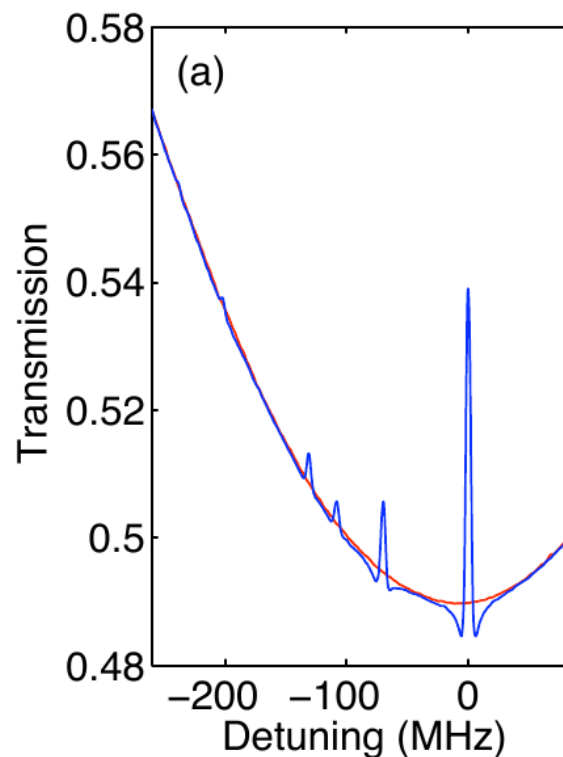
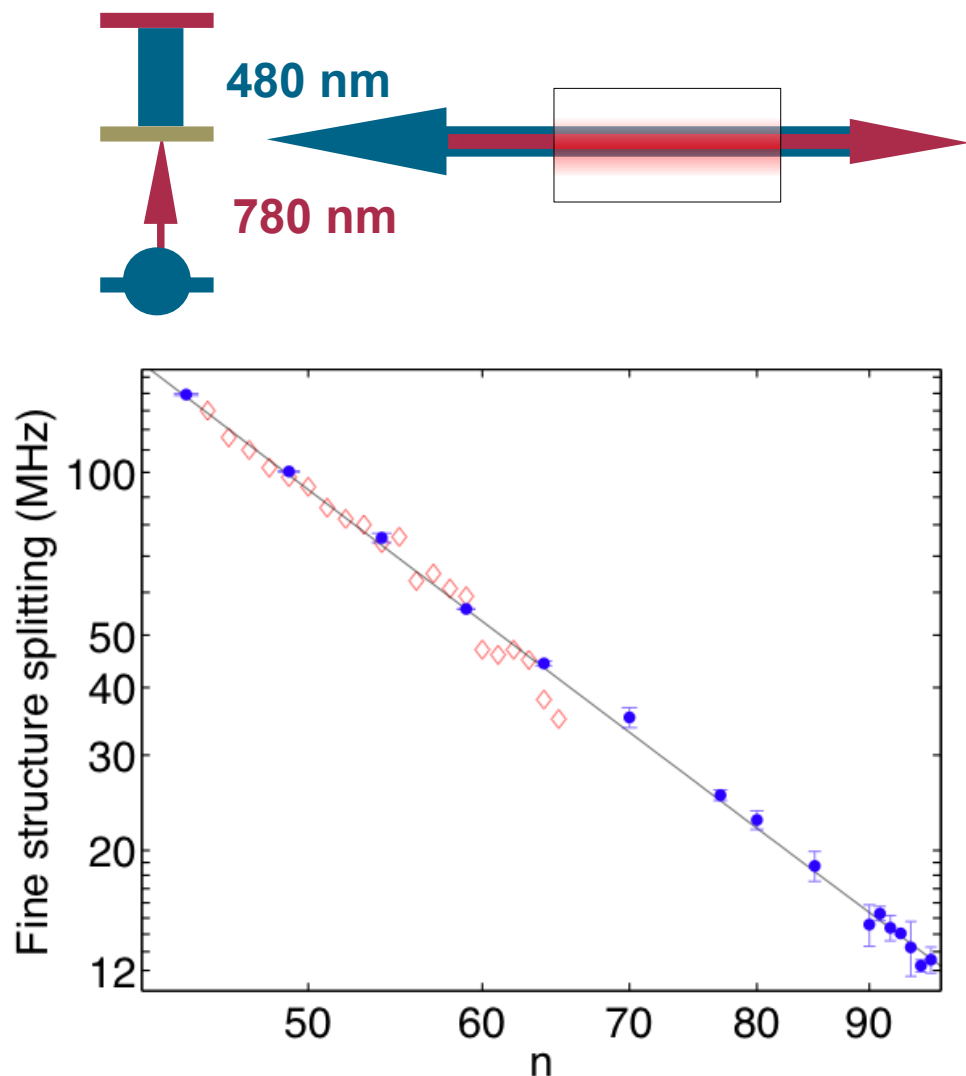
$$\mathcal{E}_{\text{at}} \sim \frac{1}{2} e / 4\pi\epsilon_0 n^4 a_0^2 \sim 5 \times 10^3 \text{ Vm}^{-1}$$

Near field  $kr < 1$  Rydberg dipoles  $r < 1 \text{ mm}$

Ground state

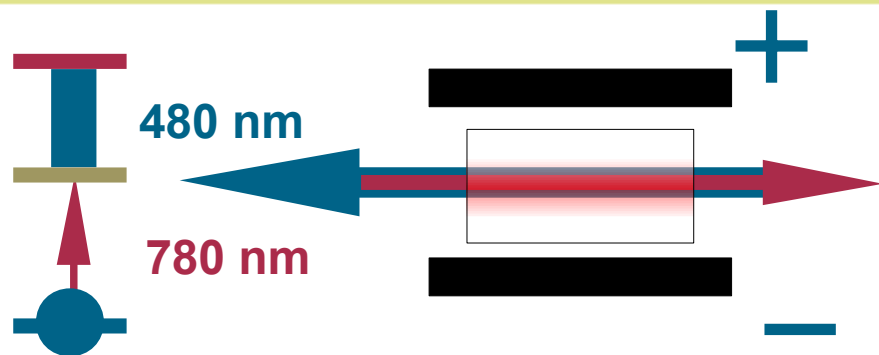




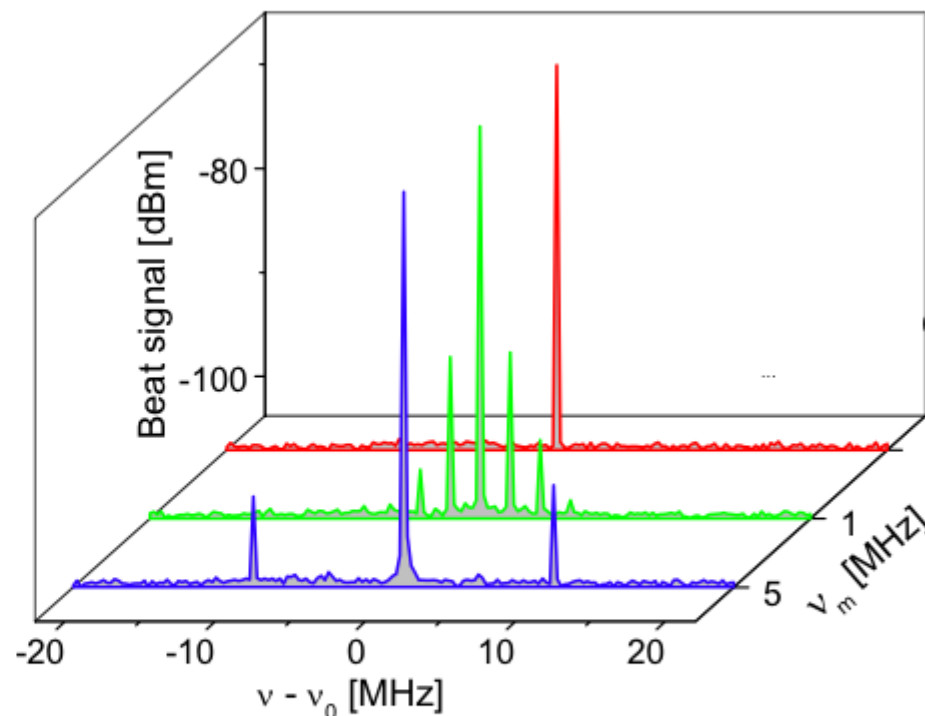


Narrow resonance – long lived coherence between ground and Rydberg states!

- ◇ K. C. Harvey and B. P. Stoicheff, *Phys. Rev. Lett.* **38**, 537 (1977).
- W. Li, I. Mourachko, M. W. Noel, and T. F. Gallagher, *Phys. Rev. A* **67**, 052502 (2003).
- A. Mohapatra, T. R. Jackson, CSA, *Phys. Rev. Lett.* **98**, 113003 (2007).



Low field electro-optic modulator

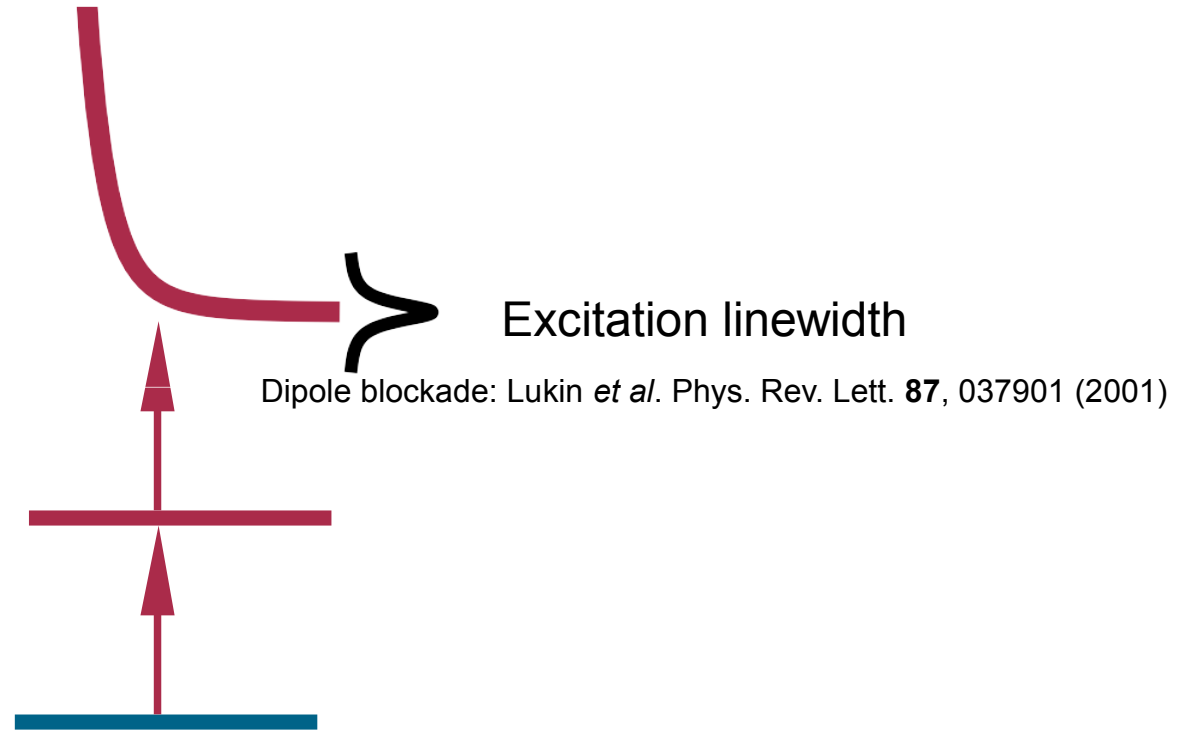
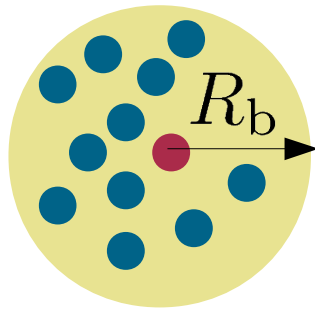


Kerr effect ( $\chi^{(3)}$ )  $10^6$  times larger than Kerr liquids (nitrobenzene)

*Giant dc Kerr effect, Mohapatra et al. Nature Phys. 4, 890 (2008).*



# What happens when the shift is larger than the linewidth?



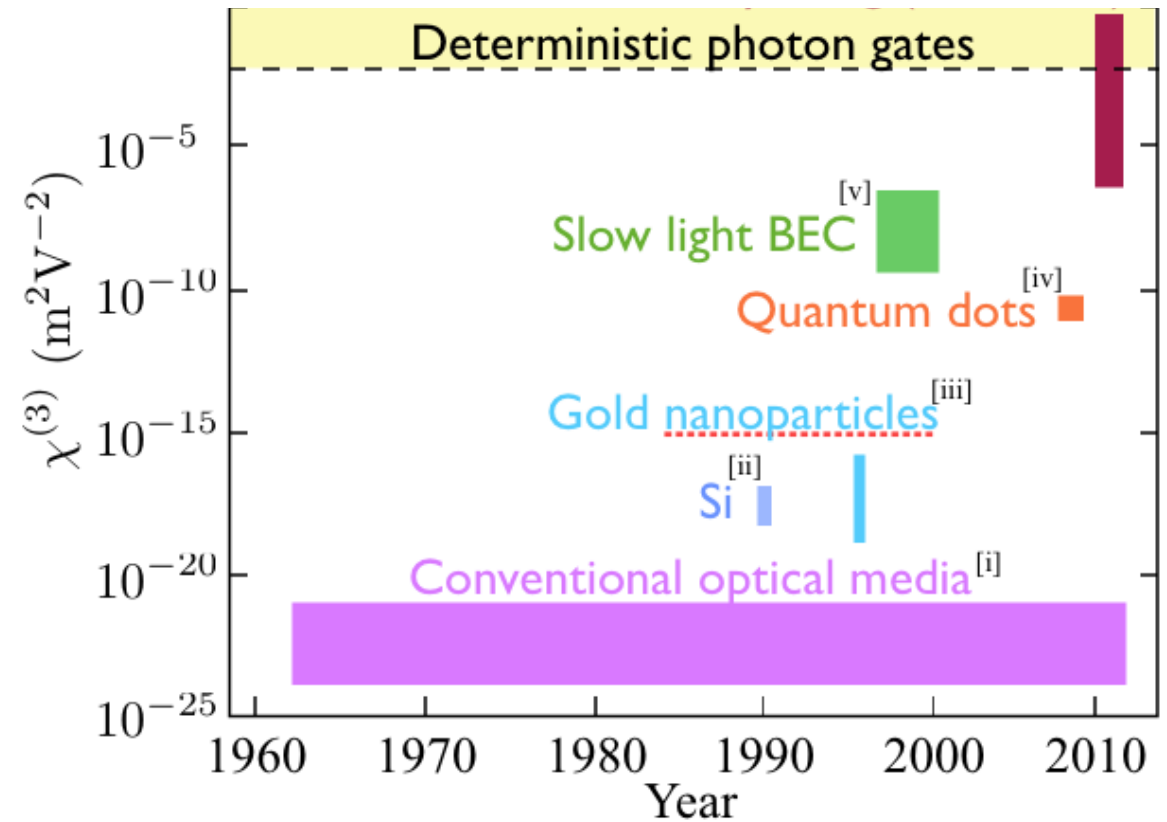
Excitation induced shift larger than the excitation linewidth

$$\chi^{(3)} = \frac{(n_g - 1)\alpha_p}{\hbar\omega}$$

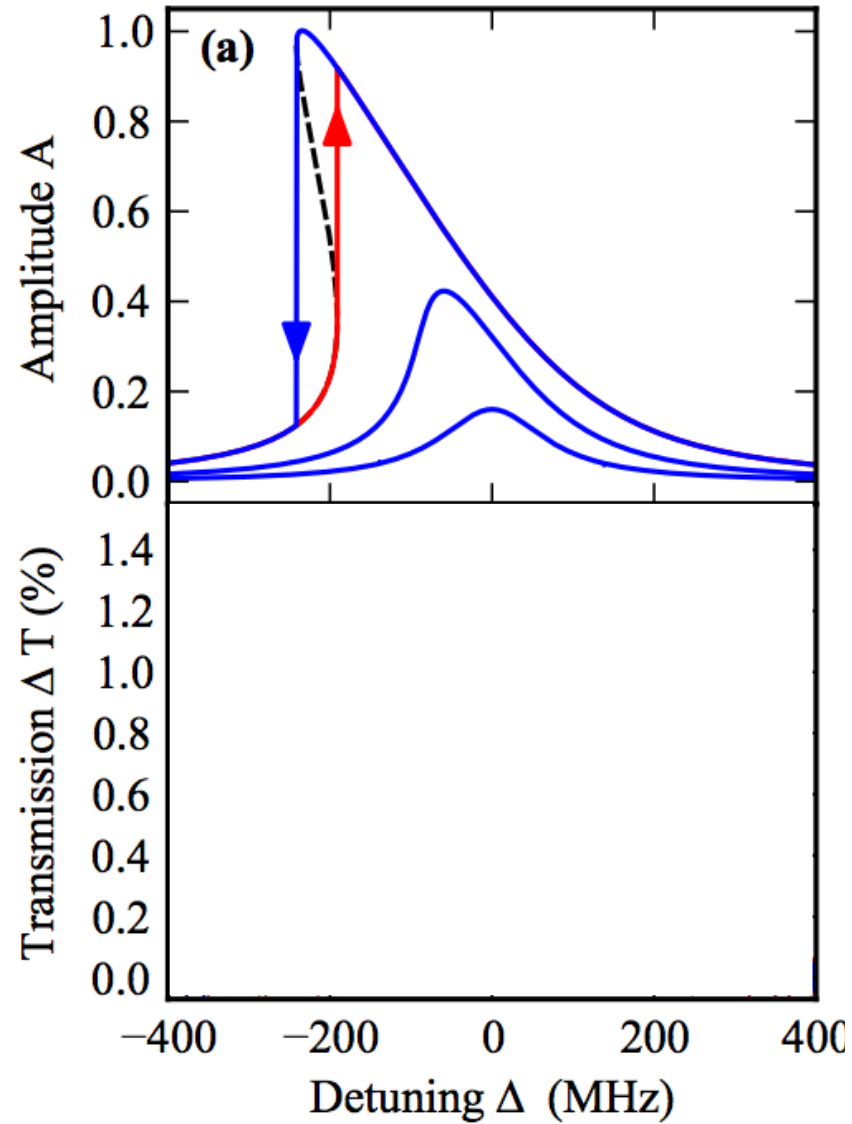
Rydberg

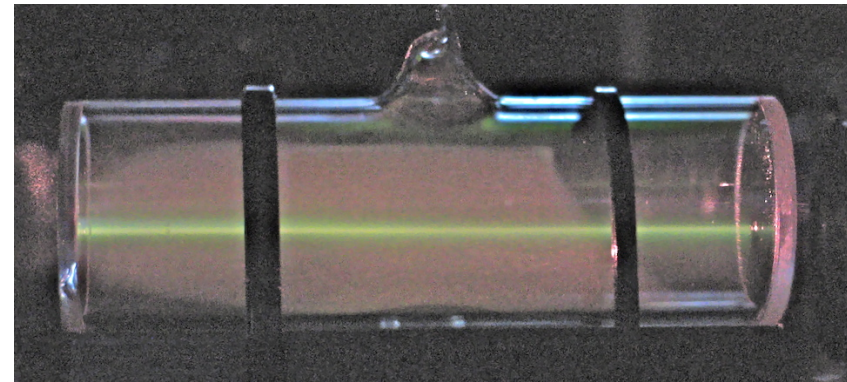
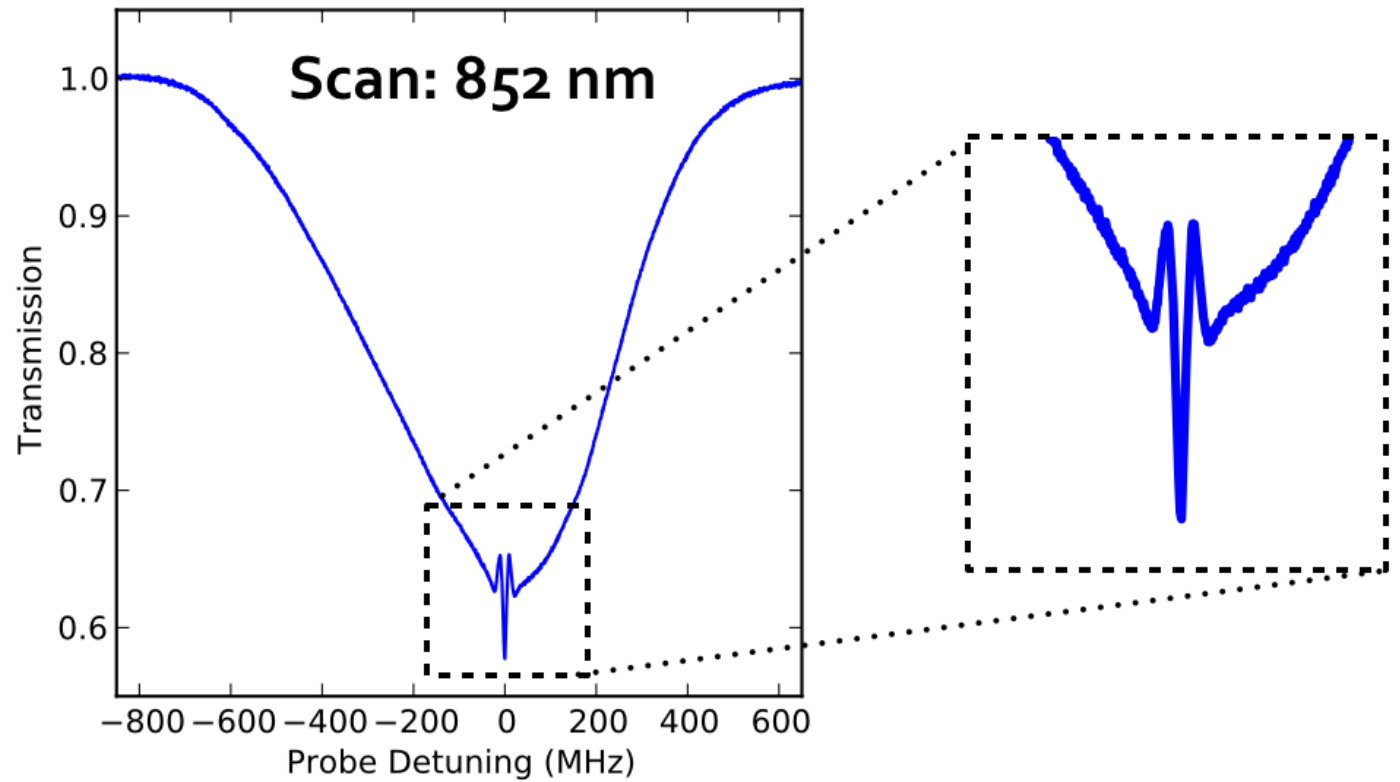
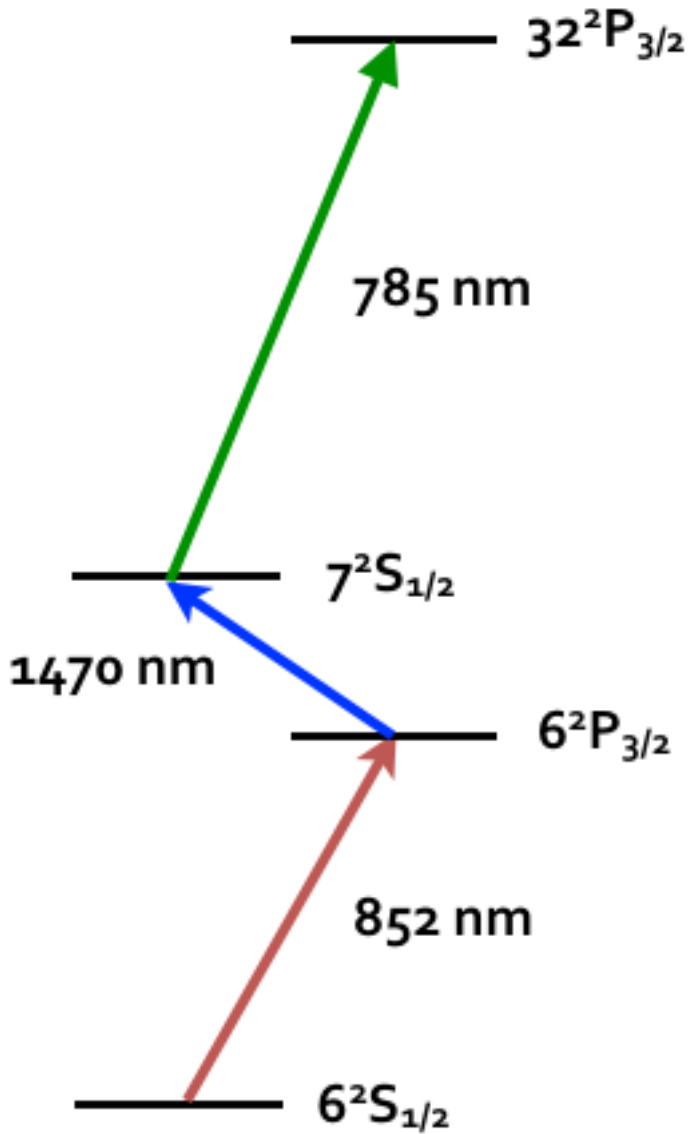
$$\chi^{(3)} \sim \mathcal{N}_b \chi^{(1)} \left( \frac{d\mathcal{E}_{ph}}{\hbar\Omega_c} \right)^2$$

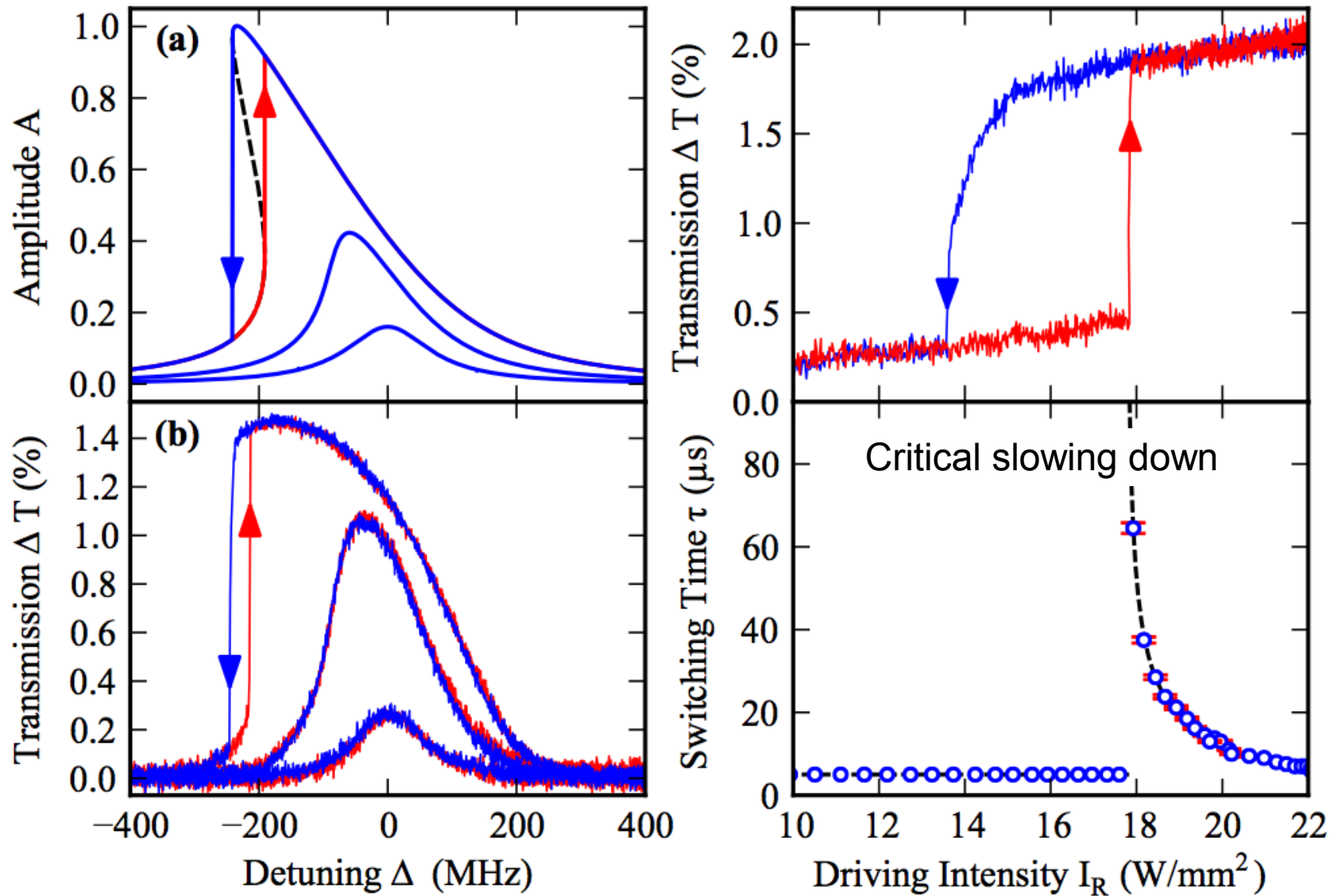
Rydberg

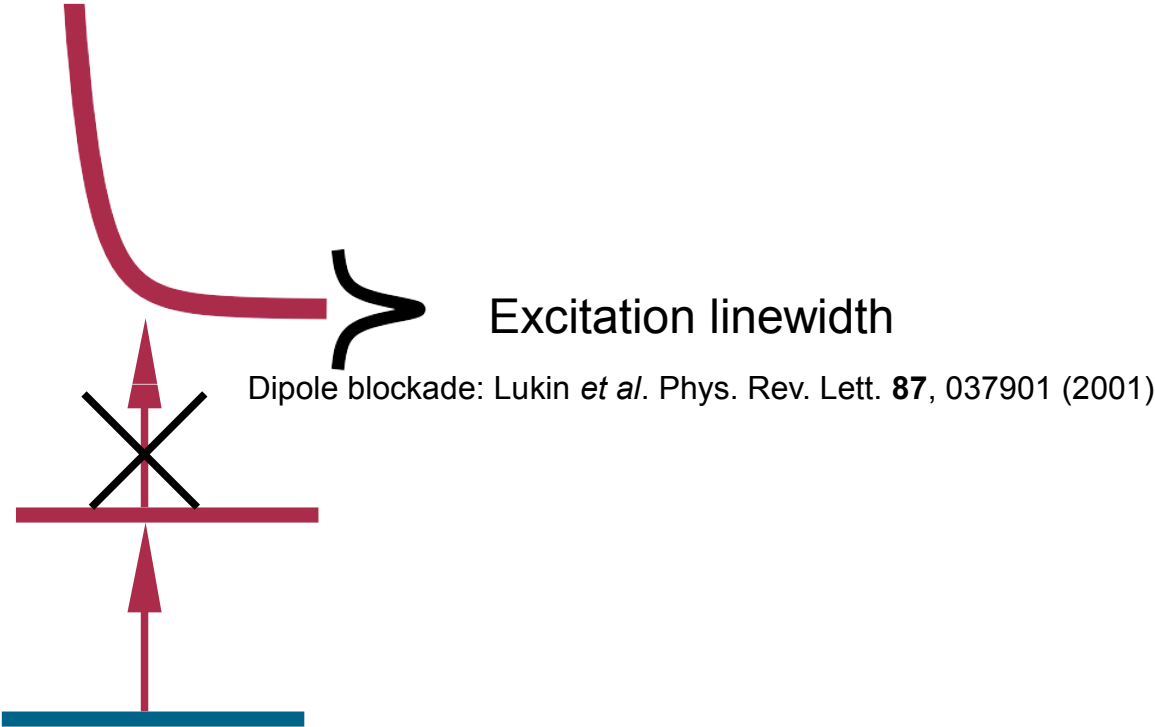
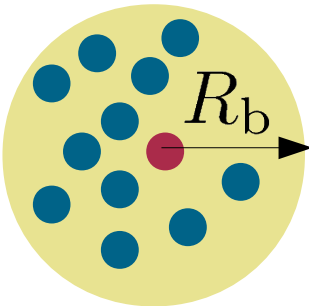


$$m\ddot{x} + \gamma\dot{x} + kx^2 + \epsilon x^3 = F \cos \omega_0 t$$



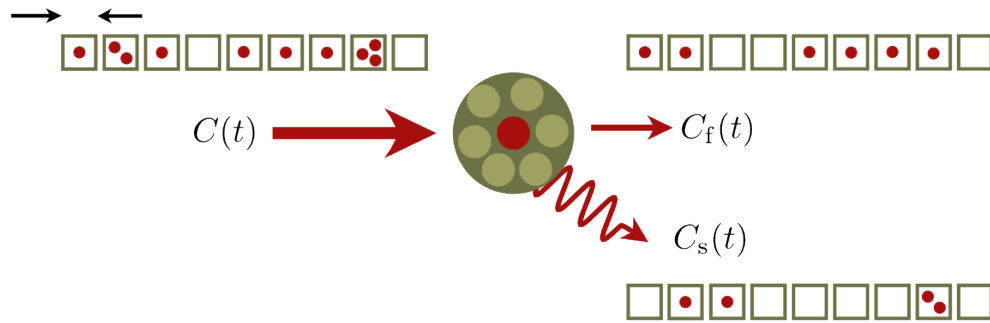




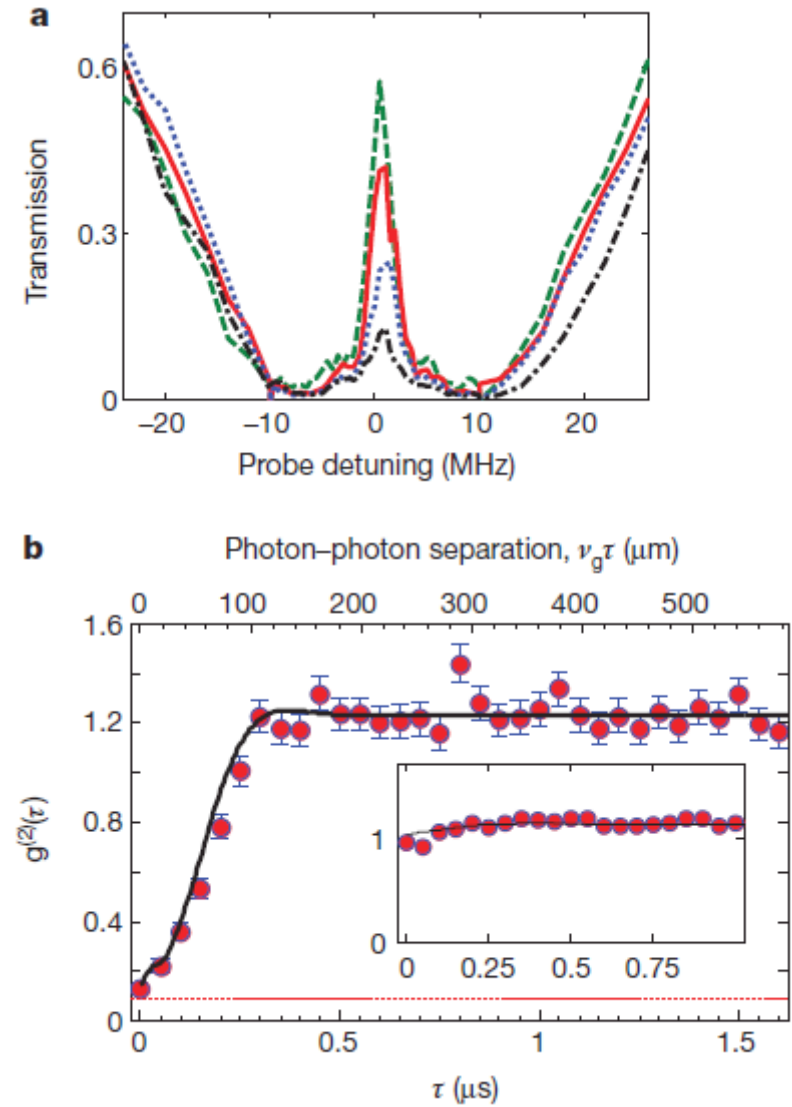


Excitation induced interaction must be larger than the excitation linewidth

Pritchard *et al.* Phys. Rev. Lett. **105**, 193603 (2010).



Peyronel *et al.* Nature 488, 57 (2012).

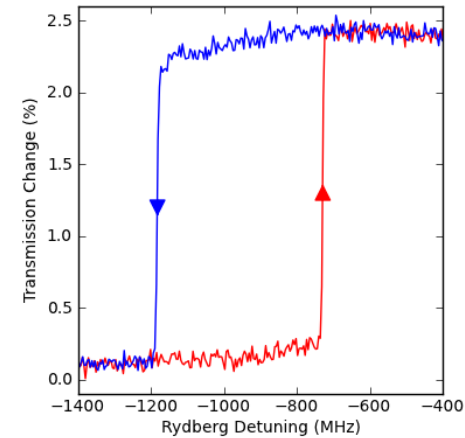


1. Typically optical non-linearities are small.

$$\chi_r^{(3)} \mathcal{E}^2 = \chi_r^{(1)} \frac{\mathcal{E}^2}{\mathcal{E}_{at}^2}$$

2. We can solve this with Rydberg atoms.

3. Optical bistability.



4. Photon-photon interactions.

