

Macroscopic quantum electrodynamics: Engineering atom–field interactions

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Outline

Introduction: quantum vacuum

Macroscopic quantum electrodynamics: a toolbox

Atom–surface interactions: enhancement by surface plasmons

Cavity QED: effective modes

Resonance energy transfer: impact of environments

Introduction:

Quantum vacuum

The quantum vacuum

“It must be that what can be spoken and thought is, for it is there for being. And there is no such thing as *nothing*.”

Parmenides

“There is no more reason for thing to exist than for *no-thing* to exist.”

Democritus



The quantum vacuum

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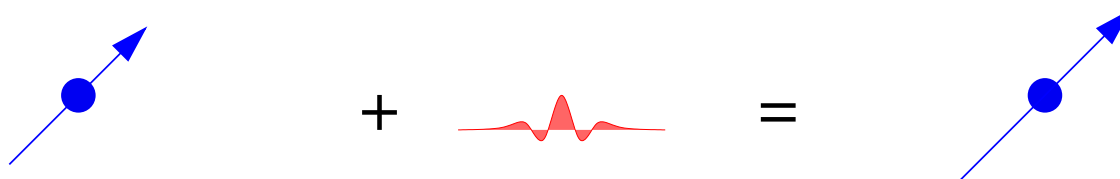
Quantum vacuum: fluctuating electromagnetic fields

Virtual photons: where do they matter?

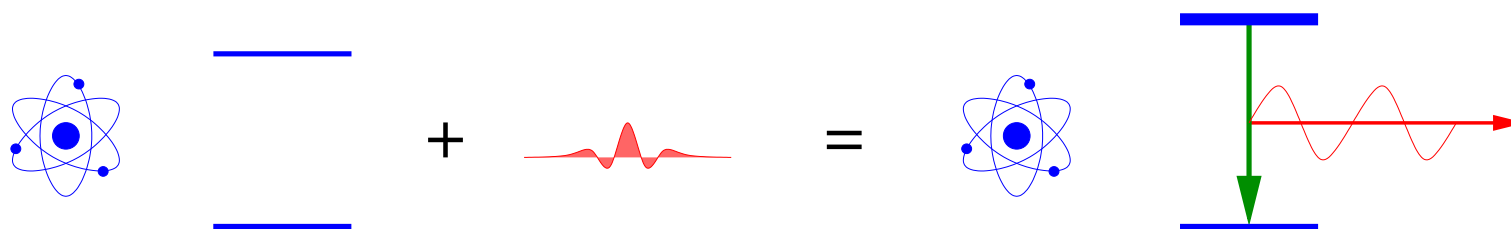
M. Brüderlin, *Japan und der Westen: Die erfüllte Leere* (Dumont, 2007);
R. Waterfield, *The First Philosophers* (Oxford University Press, 2009)

Vacuum effects in free space

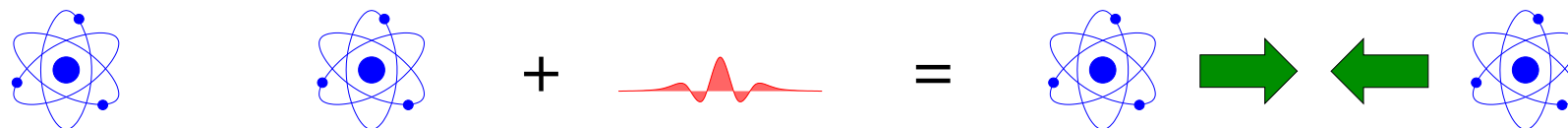
Electron: anomalous magnetic moment



Single atom: Lamb shift, spontaneous decay



Two atoms: Van der Waals force



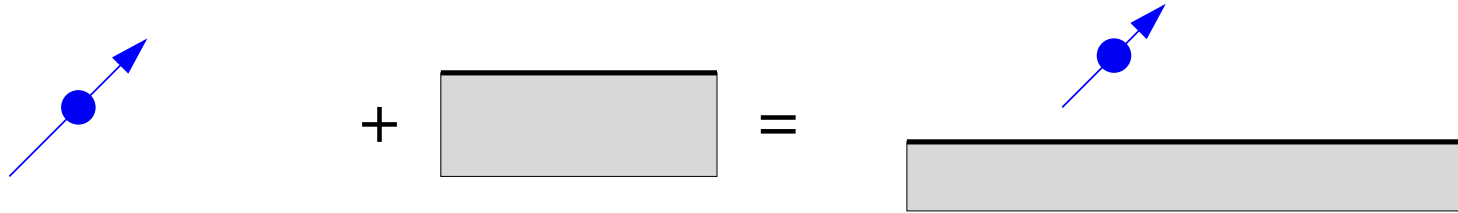
J. Schwinger, PR **73**, 416 (1948)

W. E. Lamb, R. C. Retherford, PR **72**, 241 (1947)

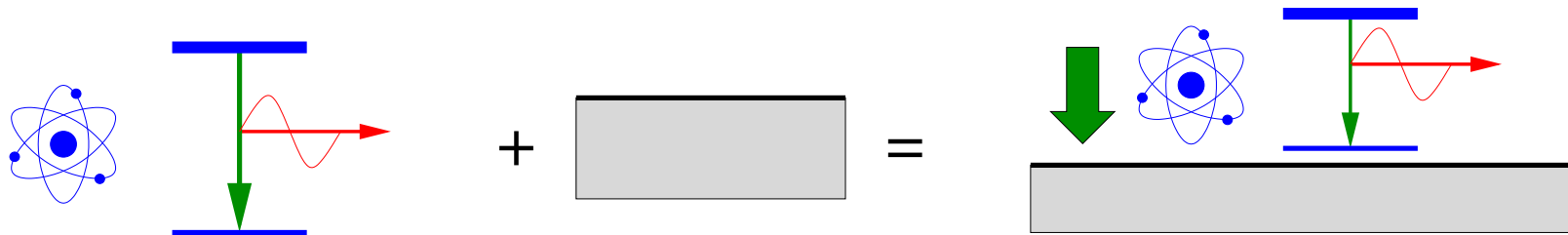
H. B. G. Casimir, D. Polder, PR **73**, 360 (1948)

Environment-affected vacuum effects

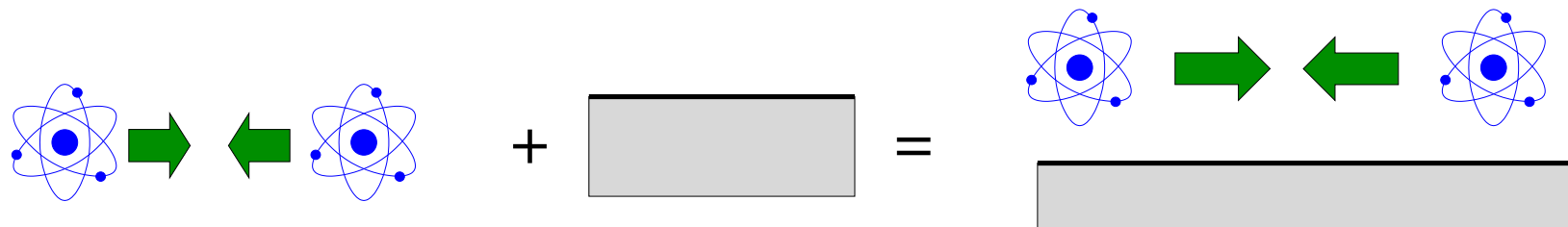
Electron: Position-dependent anomalous magnetic moment



Single atom: Casimir–Polder force, Purcell effect



Two atoms: Modified van der Waals force



G. Barton, N. S. J. Fawcett, Phys. Rep. **170**, 1 (1988); H. B. G. Casimir, D. Polder, PR **73**, 360 (1948); E. M. Purcell, PR **69**, 681 (1946); J. Mahanty, B. W. Ninham, J. Phys. A **6** 1140 (1973)

Background:

Macroscopic quantum electrodynamics

Maxwell equations + Green's tensor

Maxwell equations

$$\begin{aligned}\nabla \cdot \hat{B} &= 0 & \nabla \times \hat{E} - i\omega \hat{B} &= 0 \\ \nabla \cdot \hat{D} &= 0 & \nabla \times \hat{H} + i\omega \hat{D} &= 0\end{aligned}$$

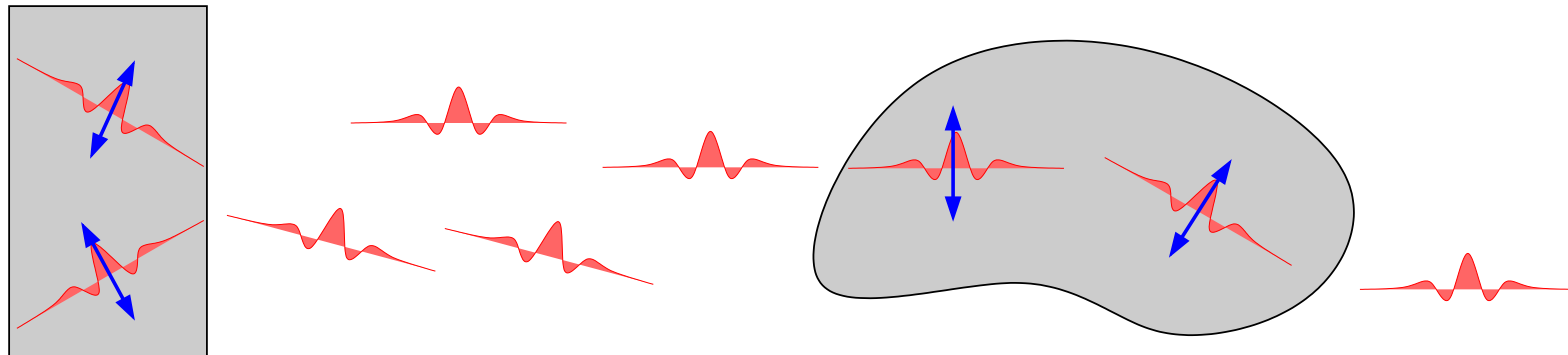
Constitutive relation

$$\hat{D} = \varepsilon_0 \varepsilon \hat{E} + \hat{P}_N$$

Electric field: $\hat{E}(\mathbf{r}) = \mu_0 \omega^2 \int d^3 r' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{P}_N(\mathbf{r}')$

Green's tensor:

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}')$$



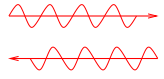
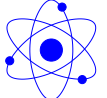
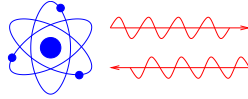
L. Knöll, S. Scheel, D.-G. Welsch, in *Coherence and Statistics of Photons and Atoms*, ed. by J. Peřina (Wiley, New York, 2001)

Quantisation + atom-field coupling

Bosonic variables: $[\hat{f}(r, \omega), \hat{f}^\dagger(r', \omega')] = \delta(r - r')\delta(\omega - \omega')$

$$\underline{\hat{P}}_N(r, \omega) = \sqrt{\frac{\hbar \epsilon_0}{\pi} \text{Im} \epsilon(r, \omega)} \hat{f}(r, \omega)$$

Hamiltonian: $\hat{H} = \hat{H}_F + \sum_A \hat{H}_A + \sum_A \hat{H}_{AF}$

- *Body-Field:* $\hat{H}_F = \int d^3r \int_0^\infty d\omega \hbar\omega \hat{f}^\dagger(r, \omega) \cdot \hat{f}(r, \omega)$ 
- *Atom:* $\hat{H}_A = \frac{\hat{p}_A^2}{2m_A} + \sum_n E_n |n\rangle \langle n|$ 
- *Electric-dipole coupling:* $\hat{H}_{AF} = -\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(r_A)$ 

Consistency with other theories

Classical electrodynamics: Maxwell eqs. + Lorentz force ✓

$$\begin{aligned}\nabla \cdot \hat{\mathbf{B}} &= 0 & \nabla \times \hat{\mathbf{E}} + \dot{\hat{\mathbf{B}}} &= \mathbf{0} \\ \nabla \cdot \hat{\mathbf{D}} &= \hat{\rho}_A & \nabla \times \hat{\mathbf{H}} - \dot{\hat{\mathbf{D}}} &= \hat{\mathbf{j}}_A \\ m_\alpha \ddot{\hat{\mathbf{r}}}_\alpha &= q_\alpha \hat{\mathbf{E}}(\hat{\mathbf{r}}_\alpha) + q_\alpha \mathcal{S}[\dot{\hat{\mathbf{r}}}_\alpha \times \hat{\mathbf{B}}(\hat{\mathbf{r}}_\alpha)]\end{aligned}$$

Statistical physics: Fluctuation–dissipation theorem ✓

$$\langle \mathcal{S}[\Delta \hat{\mathbf{E}}(\mathbf{r}, \omega) \Delta \hat{\mathbf{E}}^\dagger(\mathbf{r}', \omega')] \rangle = \frac{\hbar}{2\pi} \mu_0 \omega^2 \text{Im } \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega')$$

Free-space QED: Fundamental commutators ✓

$$[\hat{\mathbf{E}}(\mathbf{r}), \hat{\mathbf{B}}(\mathbf{r}')] = \frac{i\hbar}{\varepsilon_0} \nabla \times \delta(\mathbf{r} - \mathbf{r}') \quad \text{for } \varepsilon(\mathbf{r}, \omega) \rightarrow 1$$

Atom–surface interactions:

Enhancement by surface plasmons



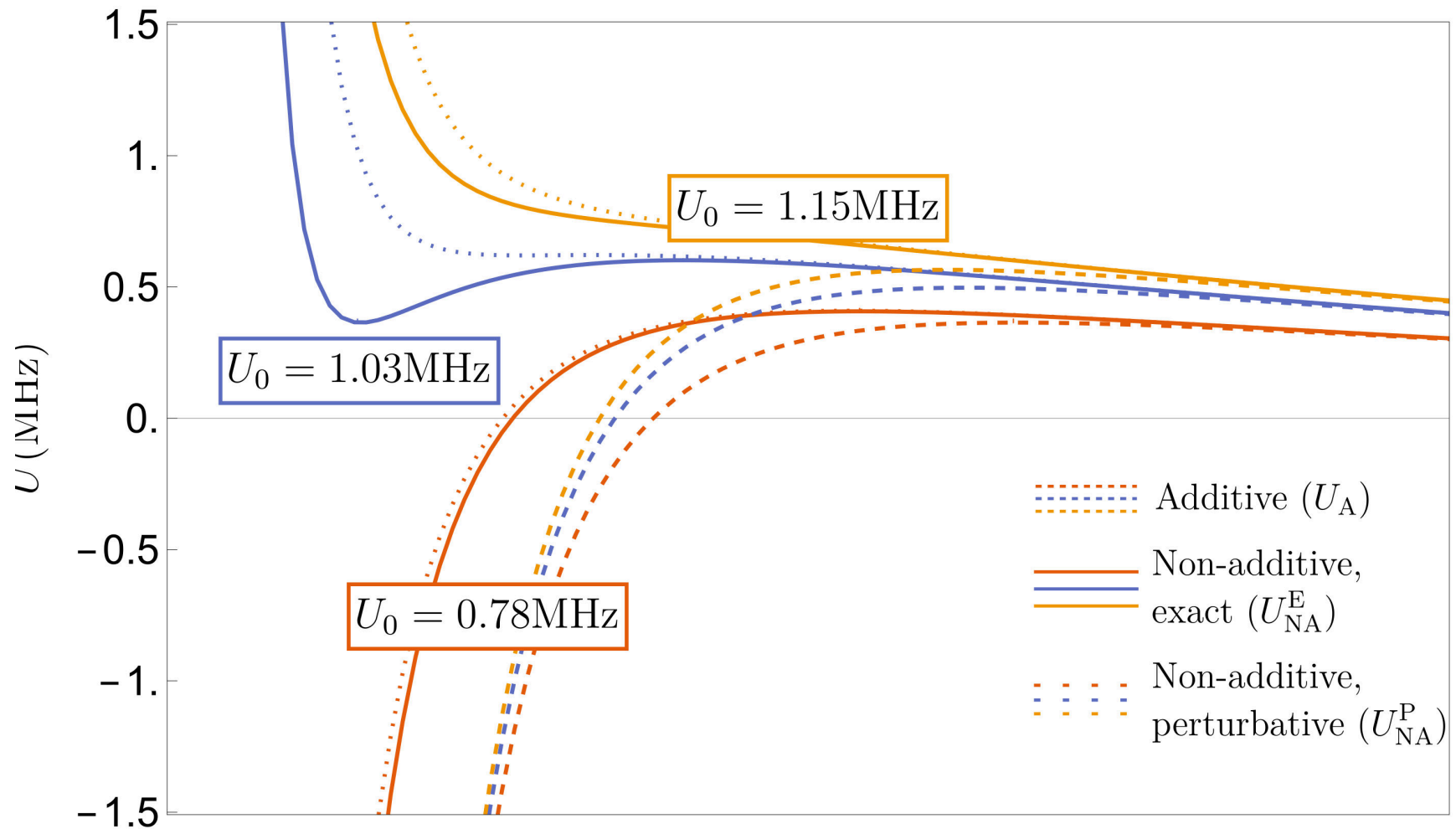
Laser-induced atom–surface interactions

Setup: atom near surface + evanescent laser

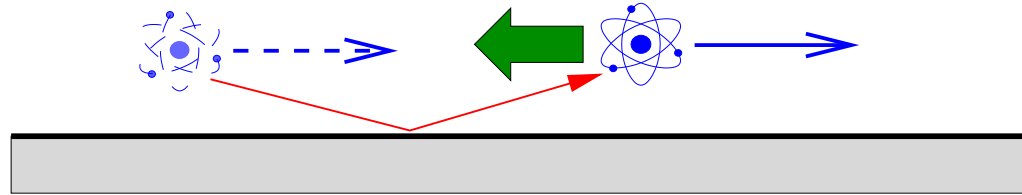
Interactions: $\hat{H}_{\text{int}} = -\hat{\mathbf{d}} \cdot \mathbf{E}_{\text{laser}}(\mathbf{r}_A) - \hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\mathbf{r}_A)$

Laser-induced atom–surface interactions

Setup: atom near surface + evanescent laser



Force on a moving atom



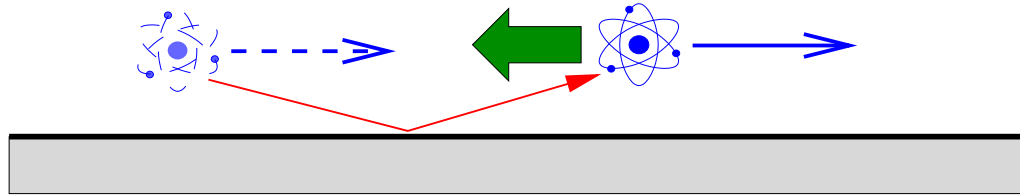
Starting point: $F = \nabla \left\langle \hat{d} \cdot [\hat{E}(r) + v \times \hat{B}(r)] \right\rangle \Big|_{r=r_A}$

Calculation: integrate coupled atom–field dynamics,
Markov approximation, linear order in v

Motion-dependent force:

- *Delay effect*: Main contribution
- *Röntgen interaction*: Relevant at large distances
- *Doppler effect*: Present for normal motion

Ground-state atom



$$F_0 = -\frac{3\omega_S v}{64\pi\epsilon_0 z_A^5} \sum_k \frac{\Gamma_k (2d_{0k}^{\perp 2} + d_{0k}^{\parallel 2})}{(\omega_{k0} + \omega_S)^3}$$

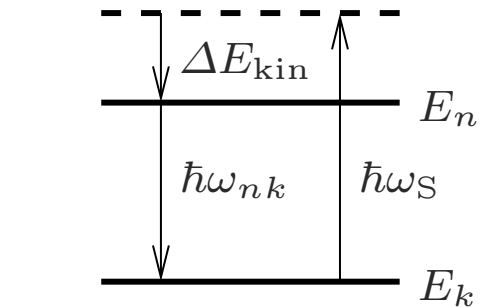
Example: Rb atom near Au surface

$$a = -v \left(1.1 \text{ s}^{-1} \right) \left[\frac{1 \text{ nm}}{z_A} \right]^8$$

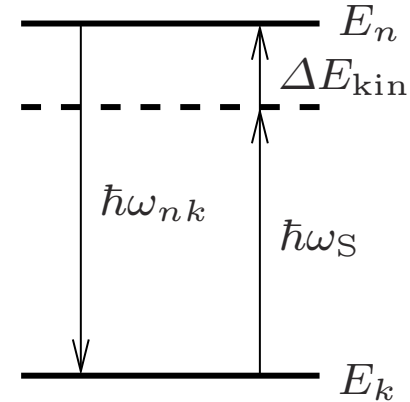
Caveat: Markov approximation breaks down

Excited Atom

(i)



(ii)

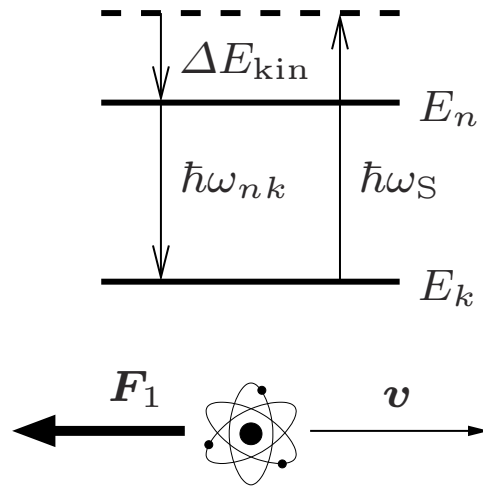


Example (i): Rb atom near Au surface

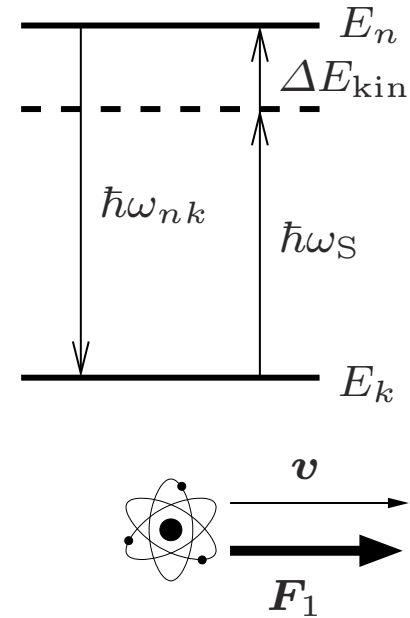
$$a = -v \left(1.1 \times 10^4 \text{ s}^{-1} \right) \left[\frac{1 \text{ nm}}{z_A} \right]^5$$

Excited Atom

(i)



(ii)



Example (ii): Cs atom near Sapphire surface

$$a = +v \left(7.1 \times 10^{11} \text{ s}^{-1} \right) \left[\frac{1 \text{ nm}}{z_A} \right]^5$$

Cavity QED:

Effective modes

Jaynes–Cummings model

Identical two-level atoms: $\hat{d}_A = d\hat{\sigma}_A + d^*\hat{\sigma}_A^\dagger$

$$\hat{H}_{AF} = \hbar \int_0^\infty d\omega g_A(\omega) [\hat{a}_A(\omega)\hat{\sigma}_A^\dagger + \hat{a}_A^\dagger(\omega)\hat{\sigma}_A] \quad (\text{rotating wave app.})$$

Effective field operators: $[\hat{a}_A(\omega), \hat{a}_B^\dagger(\omega')] = \frac{g_{AB}^2(\omega)}{g_A(\omega)g_B(\omega)}\delta(\omega - \omega')$

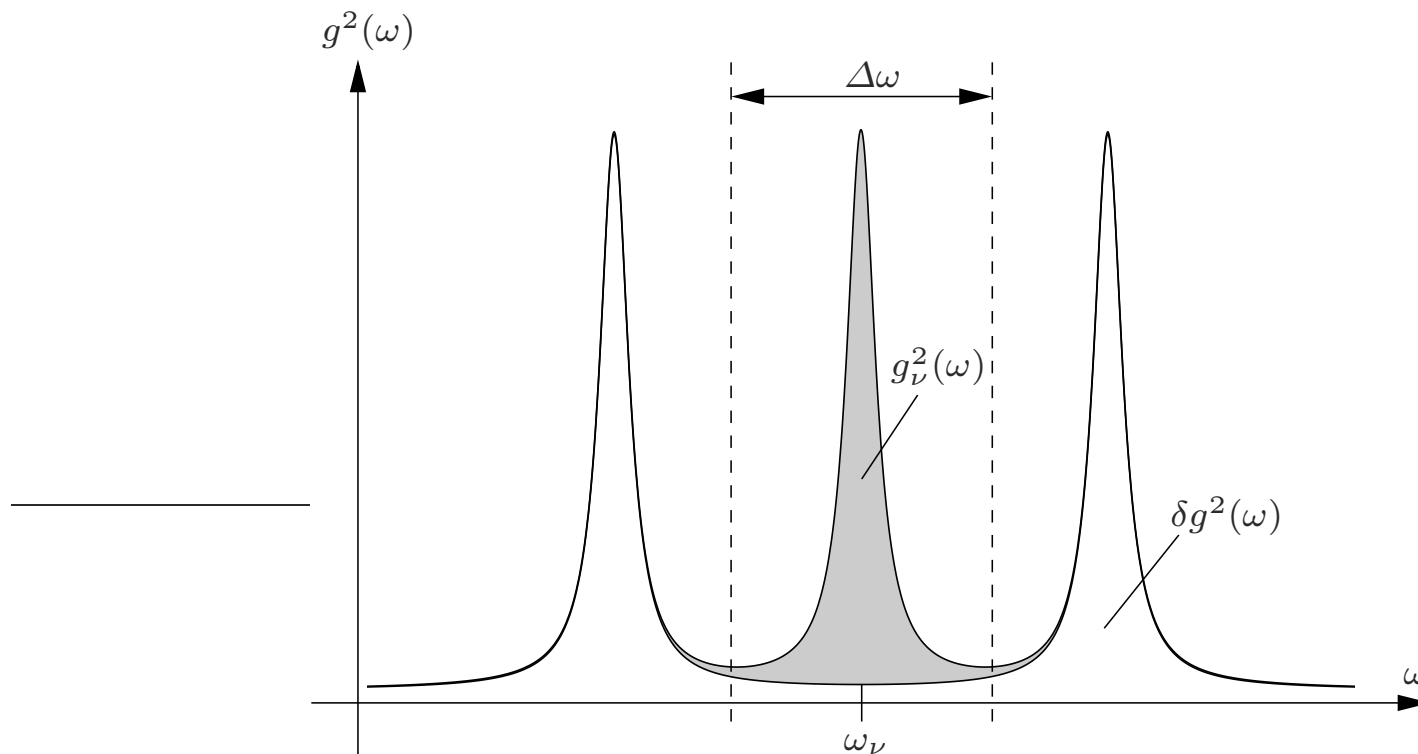
$$g_{AB}^2(\omega) = \frac{\mu_0}{\hbar\pi} \omega^2 d \cdot \text{Im } \mathbf{G}(\mathbf{r}_A, \mathbf{r}_B, \omega) \cdot d^*, \quad g_{AA}^2(\omega) \equiv g_A^2(\omega)$$

Jaynes–Cummings model

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$$\hat{H}_{AF} = \hbar \int_0^\infty d\omega g_A(\omega) [\hat{a}_A(\omega)\hat{\sigma}_A^\dagger + \hat{a}_A^\dagger(\omega)\hat{\sigma}_A] \quad (\text{rotating wave app.})$$

Single-mode approximation: $g_{AB}^2(\omega) = g_{AB}^2(\omega_\nu) \frac{\gamma_\nu^2/4}{(\omega - \omega_\nu)^2 + \gamma_\nu^2/4}$



Strong coupling: two atoms

Hamiltonian: basis states $|+\rangle|0_\nu\rangle$, $|0_A0_B\rangle|1_\nu\rangle$, $|-\rangle|0_\nu\rangle$

$$\hat{H} = \frac{\hbar}{2} \begin{pmatrix} 0 & \sqrt{\gamma_\nu \pi N} & 0 \\ \sqrt{\gamma_\nu \pi N} & \Delta & \sqrt{\frac{\gamma_\nu \pi}{N}} [g_A^2(\omega_\nu) - g_B^2(\omega_\nu)] \\ 0 & \sqrt{\frac{\gamma_\nu \pi}{N}} [g_A^2(\omega_\nu) - g_B^2(\omega_\nu)] & 0 \end{pmatrix}$$

$$N = g_A^2(\omega_\nu) + g_B^2(\omega_\nu) + 2g_{AB}^2(\omega_\nu), \quad \Delta = \omega_\nu - \omega_A$$

Strong coupling: two atoms

Hamiltonian: basis states $|+\rangle|0_\nu\rangle$, $|0_A0_B\rangle|1_\nu\rangle$, $|-\rangle|0_\nu\rangle$

$$\hat{H} = \frac{\hbar}{2} \begin{pmatrix} 0 & \sqrt{\gamma_\nu \pi N} & 0 \\ \sqrt{\gamma_\nu \pi N} & \Delta & \sqrt{\frac{\gamma_\nu \pi}{N}} [g_A^2(\omega_\nu) - g_B^2(\omega_\nu)] \\ 0 & \sqrt{\frac{\gamma_\nu \pi}{N}} [g_A^2(\omega_\nu) - g_B^2(\omega_\nu)] & 0 \end{pmatrix}$$

$$N = g_A^2(\omega_\nu) + g_B^2(\omega_\nu) + 2g_{AB}^2(\omega_\nu), \quad \Delta = \omega_\nu - \omega_A$$

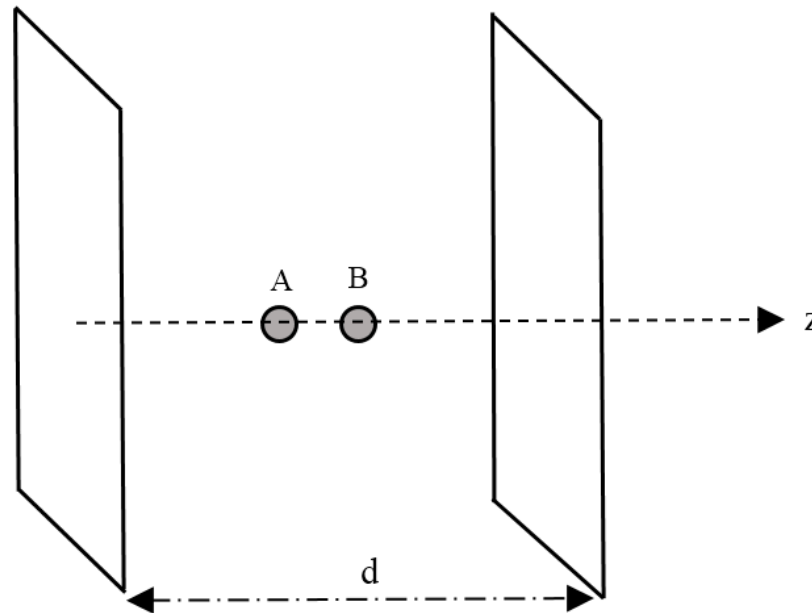
Eigenenergies: $E_\pm = \frac{\hbar}{2} \Delta \pm \frac{\hbar}{2} \sqrt{\Omega^2(r_A, r_B) + \Delta^2}$, $E_0 = 0$

$$\Omega^2(r_A, r_B) = 2\gamma_\nu \pi \frac{[g_A^2(\omega_\nu) + g_{AB}^2(\omega_\nu)]^2 + [g_B^2(\omega_\nu) + g_{AB}^2(\omega_\nu)]^2}{g_A^2(\omega_\nu) + g_B^2(\omega_\nu) + 2g_{AB}^2(\omega_\nu)}$$

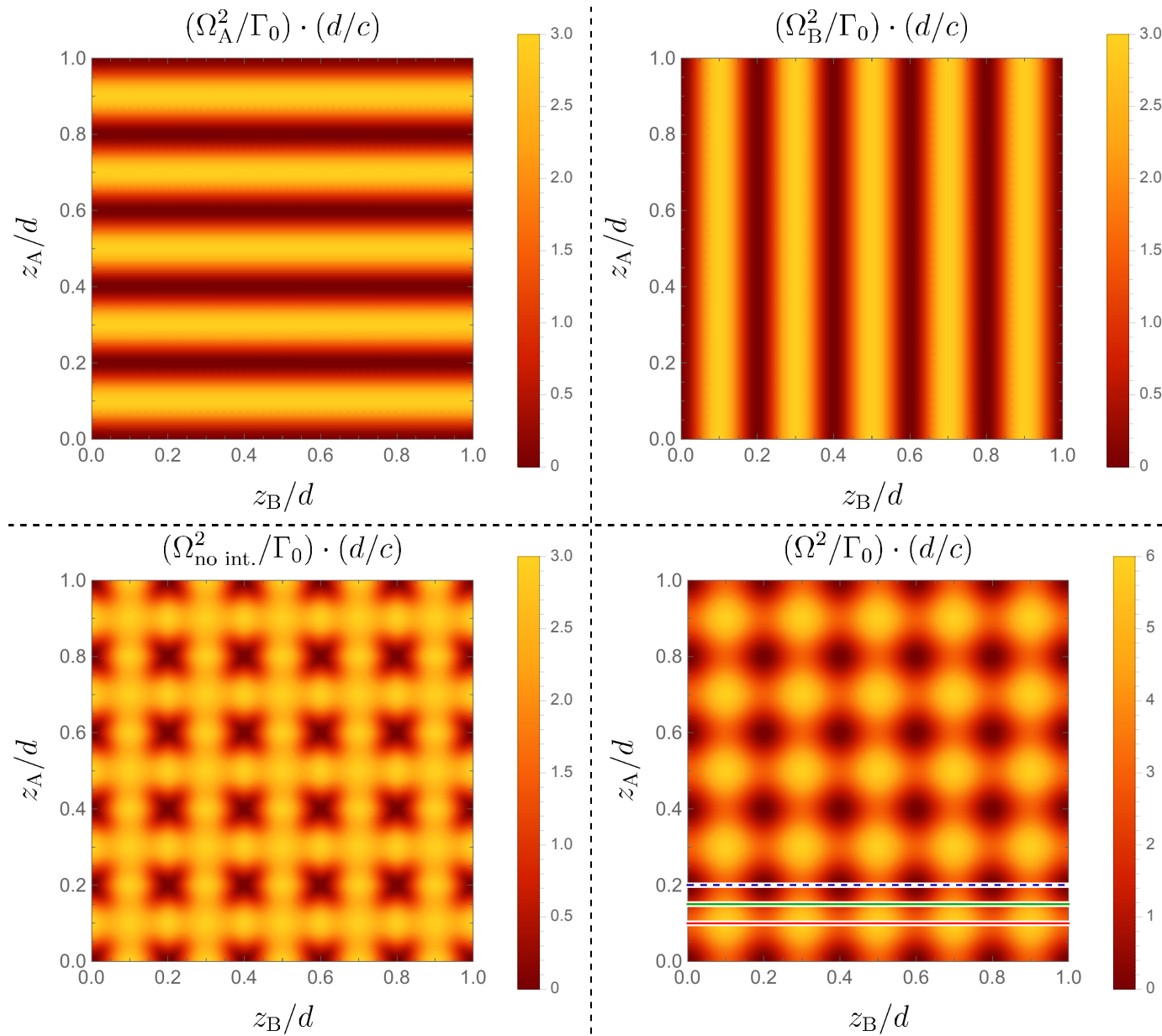
Example: Planar cavity

Two perfect mirrors: $r_p = -r_s = 1 - \delta \Rightarrow \gamma_\nu = 2c\delta/d$

$$\gamma_\nu \pi g_{AB}^2(\omega_\nu) = \frac{3c\Gamma}{8d} \left\{ \cos \left[\frac{(2d - z_A - z_B)\omega_\nu}{c} \right] - \cos \left[\frac{(2d + z_A - z_B)\omega_\nu}{c} \right] \right. \\ \left. - \cos \left[\frac{(2d - z_A + z_B)\omega_\nu}{c} \right] - \cos \left[\frac{(z_A + z_B)\omega_\nu}{c} \right] \right\}$$



Example: Planar cavity



Photonic Bose–Einstein condensate

Normal Bose–Einstein condensate: atoms in a trap

- *Conserved number of particles*
- *Thermalisation by collisions*
- *Cooling below critical temperature*

⇒ **Macroscopic occupation of ground state**

Photonic Bose–Einstein condensate

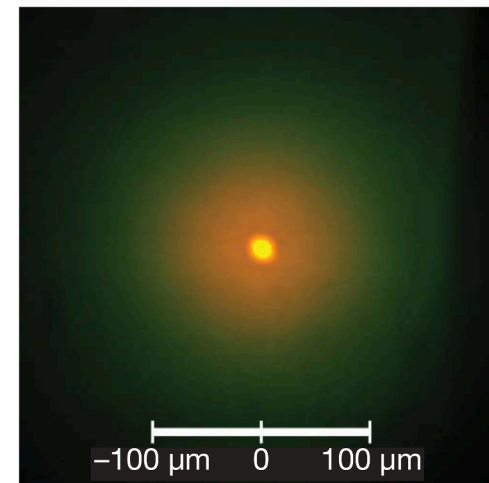
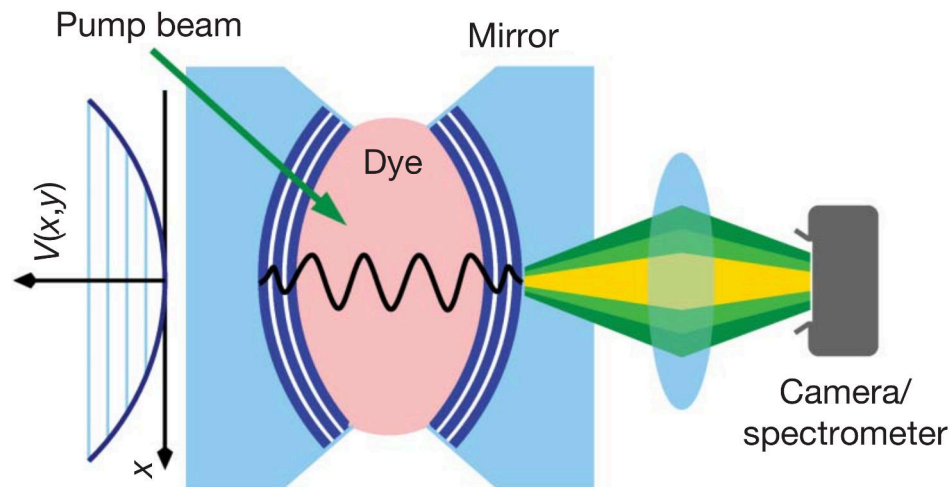
Photonic Bose–Einstein condensate: photons

- *Conserved number of particles?*
- *Thermalisation by collisions?*
- *Cooling below critical temperature?*

Photonic Bose–Einstein condensate

Photonic Bose–Einstein condensate: photons + dye in cavity

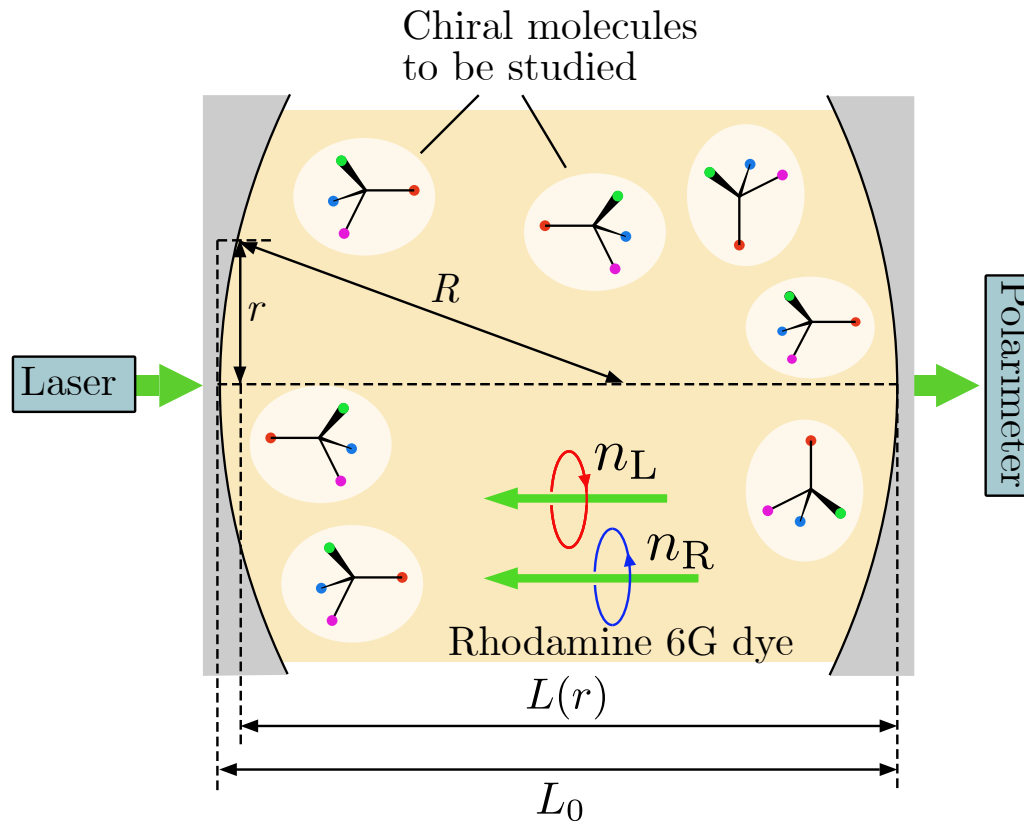
- *Conserved number of particles:* driving laser
- *Thermalisation by collisions:* absorption/emission by dye molecules
- *Cooling below critical temperature:* driving beyond threshold



⇒ **Macroscopic occupation of lowest-energy mode**

J. Klaers, J. Schmitt, F. Vewinger, M. Weitz, *Nature* **468**, 545 (2010)

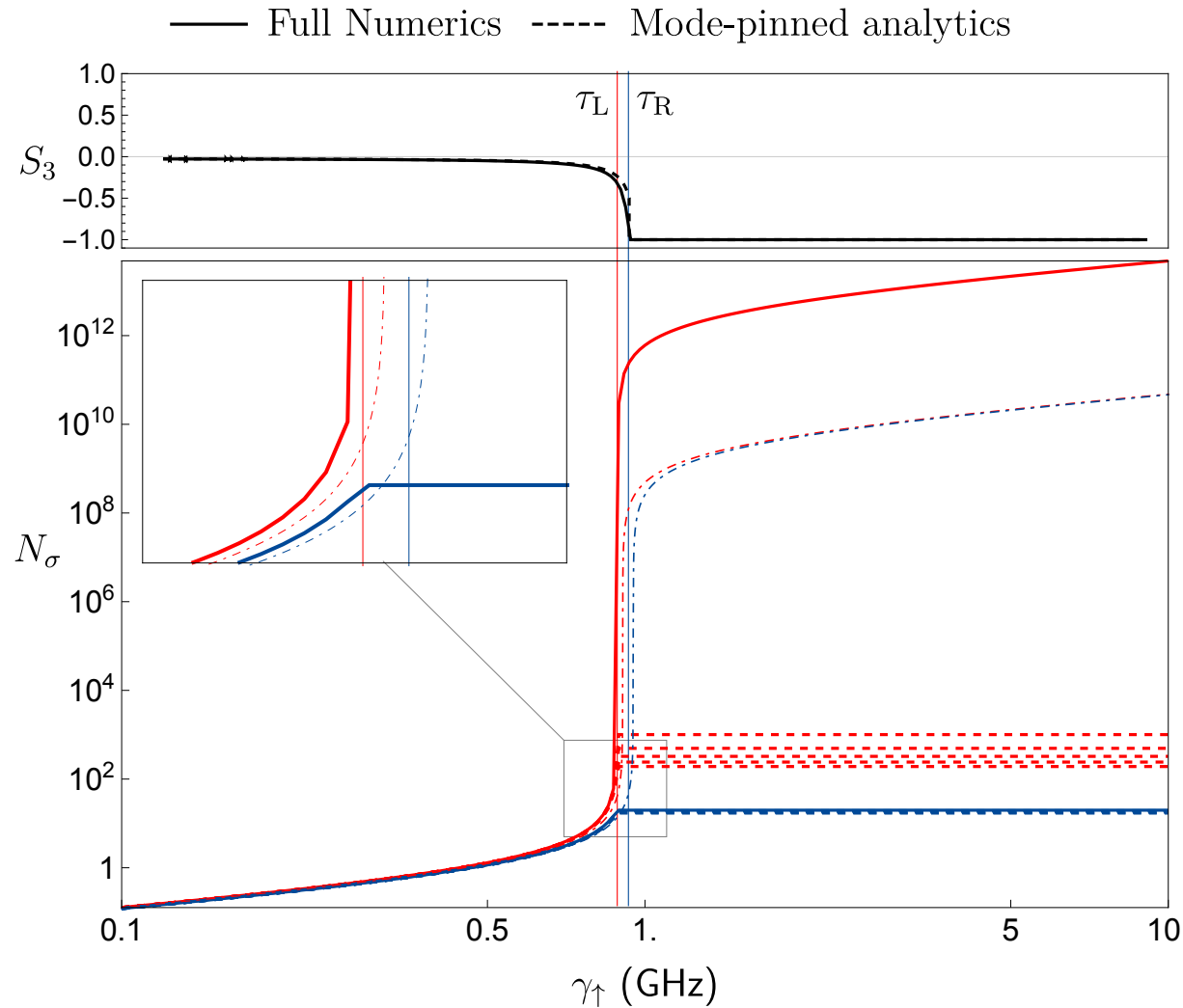
Symmetry breaking: setup and dynamics



$$\dot{N}_\nu = \kappa N_\nu - \gamma_{\uparrow\nu} N_\nu M(1 - p_e) + \gamma_{\downarrow\nu} (N_\nu + 1) M p_e$$

$$\begin{aligned} \dot{p}_e = & - \left[\gamma_{\downarrow} + \sum_{\nu} (l + 1) (N_\nu + 1) \gamma_{\downarrow\nu} \right] p_e \\ & + \left[\gamma_{\uparrow} + \sum_{\nu} (l + 1) N_\nu \gamma_{\uparrow\nu} \right] (1 - p_e) \end{aligned}$$

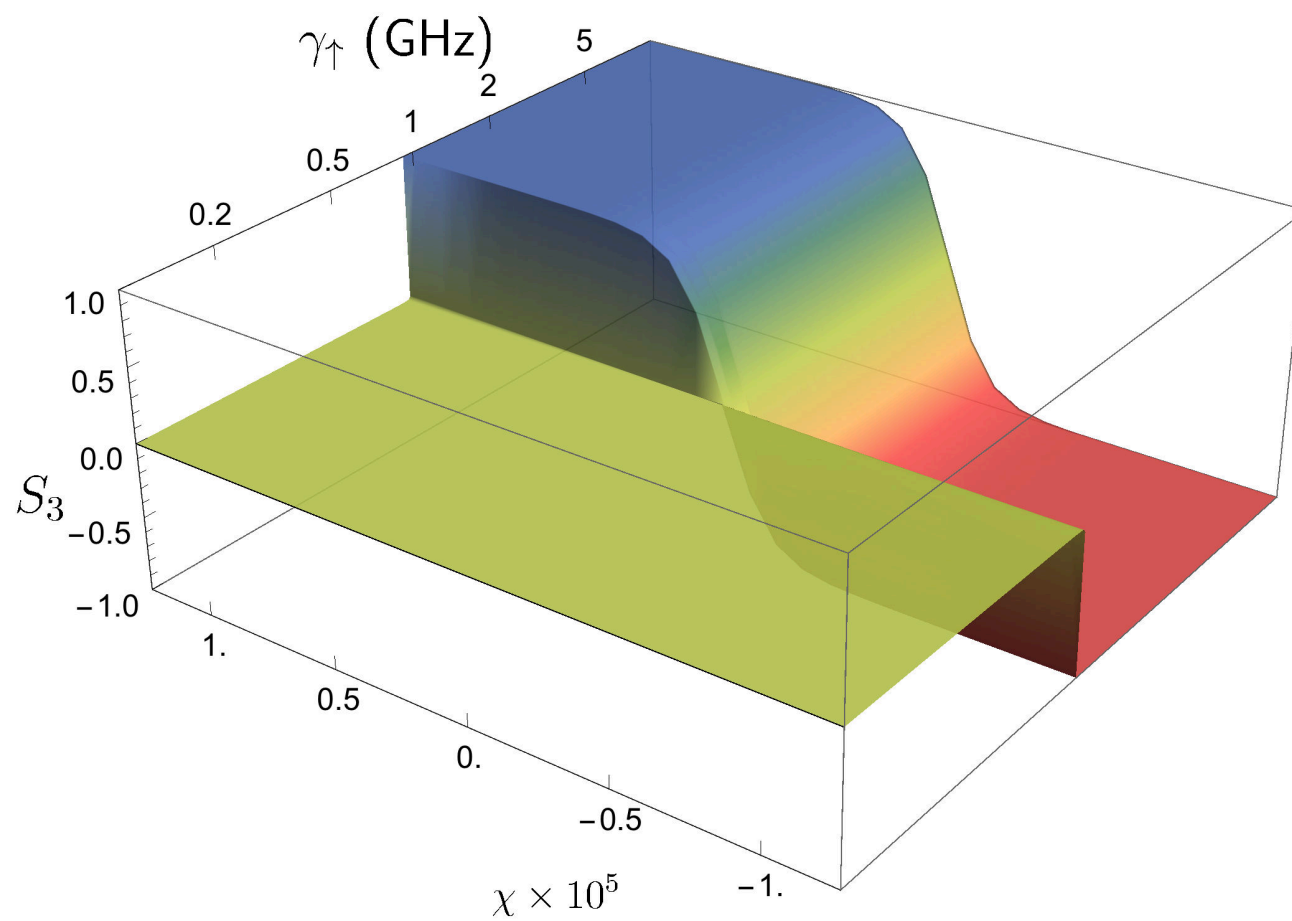
Driven-dissipative equilibrium



— Ground L mode - - - - Other L modes - - - - Single L mode
 — Ground R mode - - - - Other R modes - - - - Single R mode

Detecting enantiomeric excess

Stokes parameter: $S_3 = \frac{N_R - N_L}{N_R + N_L}$



Resonance energy transfer:

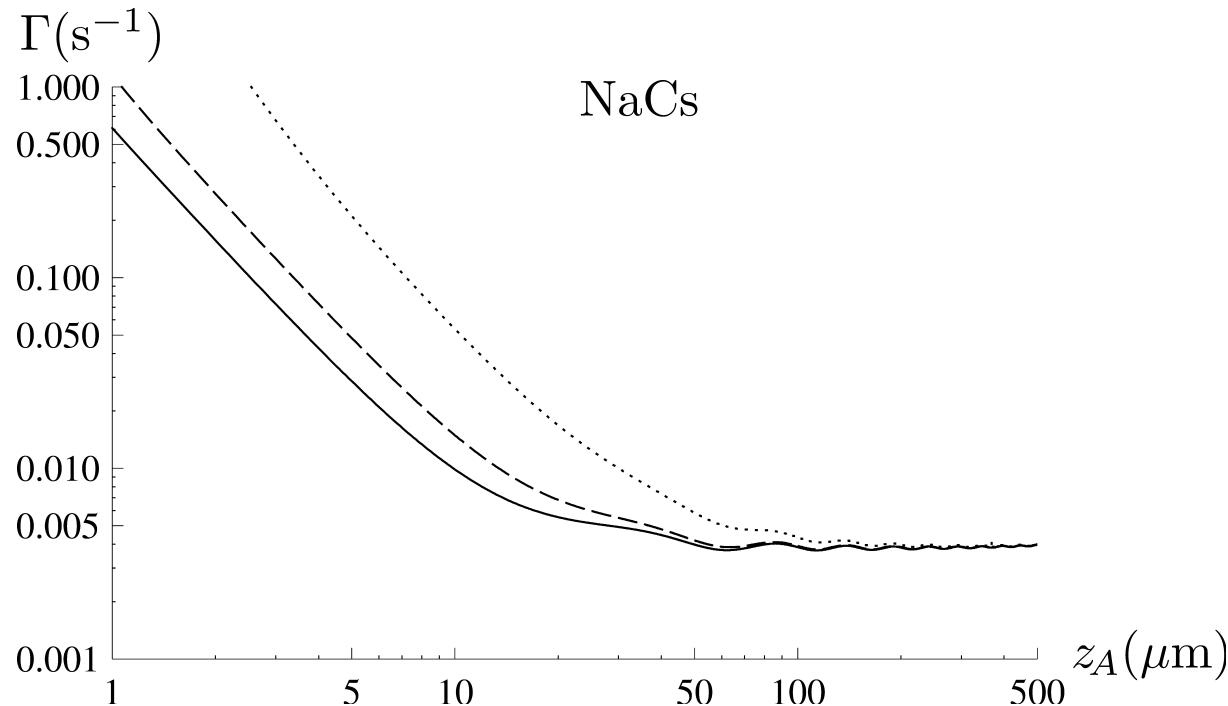
Impact of environments

The Purcell effect

Fermi's golden rule: $\Gamma = \frac{2\pi}{\hbar} \sum_f |M_{fi}|^2 \delta(E_i - E_f)$

Spontaneous decay: $M_{fi} = \langle 1_\lambda(\mathbf{r}, \omega) | \langle k_A | -\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}(\mathbf{r}_A) | n_A \rangle | \{0\} \rangle$

$$\Gamma_n = \sum_{k < n} \Gamma_{n \rightarrow k} = \frac{2\mu_0}{\hbar} \sum_{k < n} \omega_{nk}^2 \mathbf{d}_{nk} \cdot \text{Im} \mathbf{G}(\mathbf{r}_A, \mathbf{r}_A, \omega_{nk}) \cdot \mathbf{d}_{kn}$$



Interatomic Coulombic decay

Process:

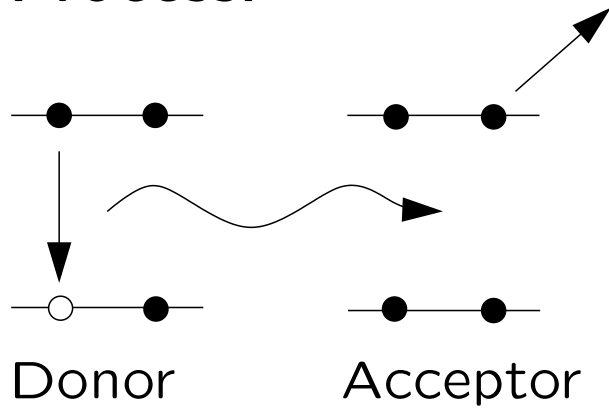
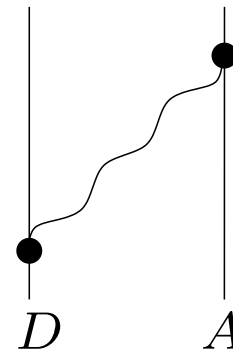


Diagram:



Interatomic Coulombic decay

Process:

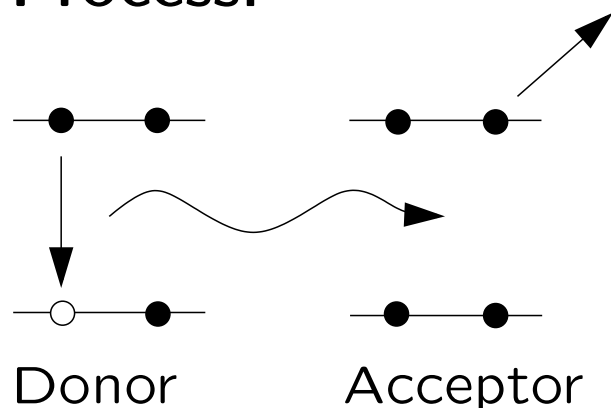
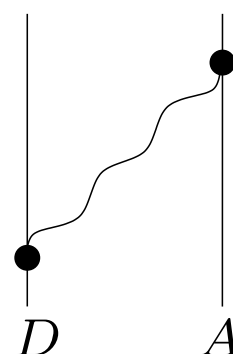


Diagram:



Fermi's golden rule: $\Gamma = \frac{2\pi}{\hbar} \sum_f |M_{fi}|^2 \delta(E_i - E_f)$

Matrix element: $M_{fi} = \sum_{\psi} \frac{\langle f | \hat{H}_{\text{int}} | \psi \rangle \langle \psi | \hat{H}_{\text{int}} | i \rangle}{E_i - E_{\psi} + i\epsilon}$

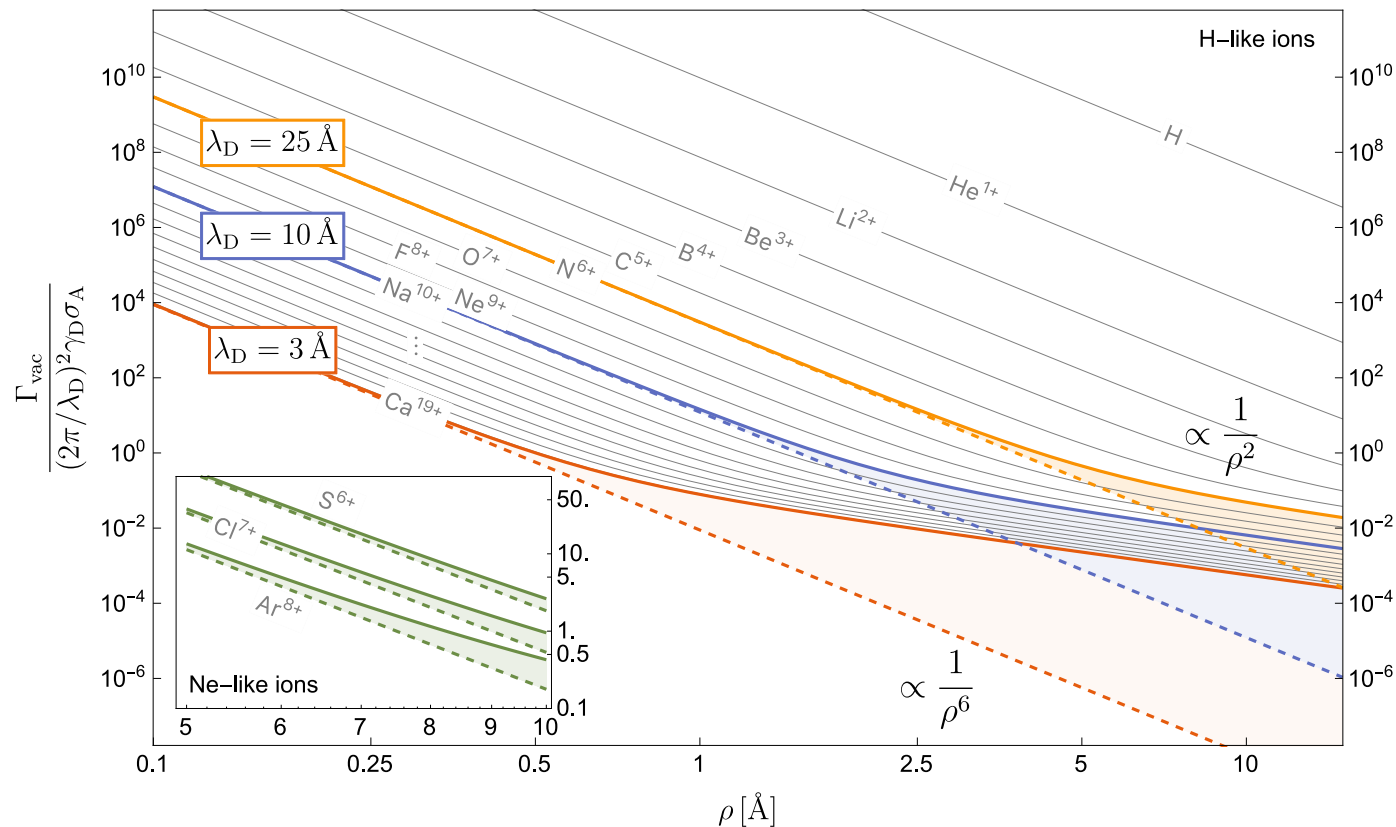
$$\Gamma = 2\pi^2 \sum_{k < n} \Gamma_{n \rightarrow k}^D \sigma_A(\hbar\omega_{nk}) \text{tr}[\mathbf{G}(\mathbf{r}_D, \mathbf{r}_A, \omega_{nk}) \cdot \mathbf{G}^*(\mathbf{r}_A, \mathbf{r}_D, \omega_{nk})]$$

L. S. Cederbaum *et al.*, PRL **79**, 4478 (1997);

J. L. Hemmerich, R. Bennett and S.Y.B., Nature Commun. **9**, 2934 (2018)

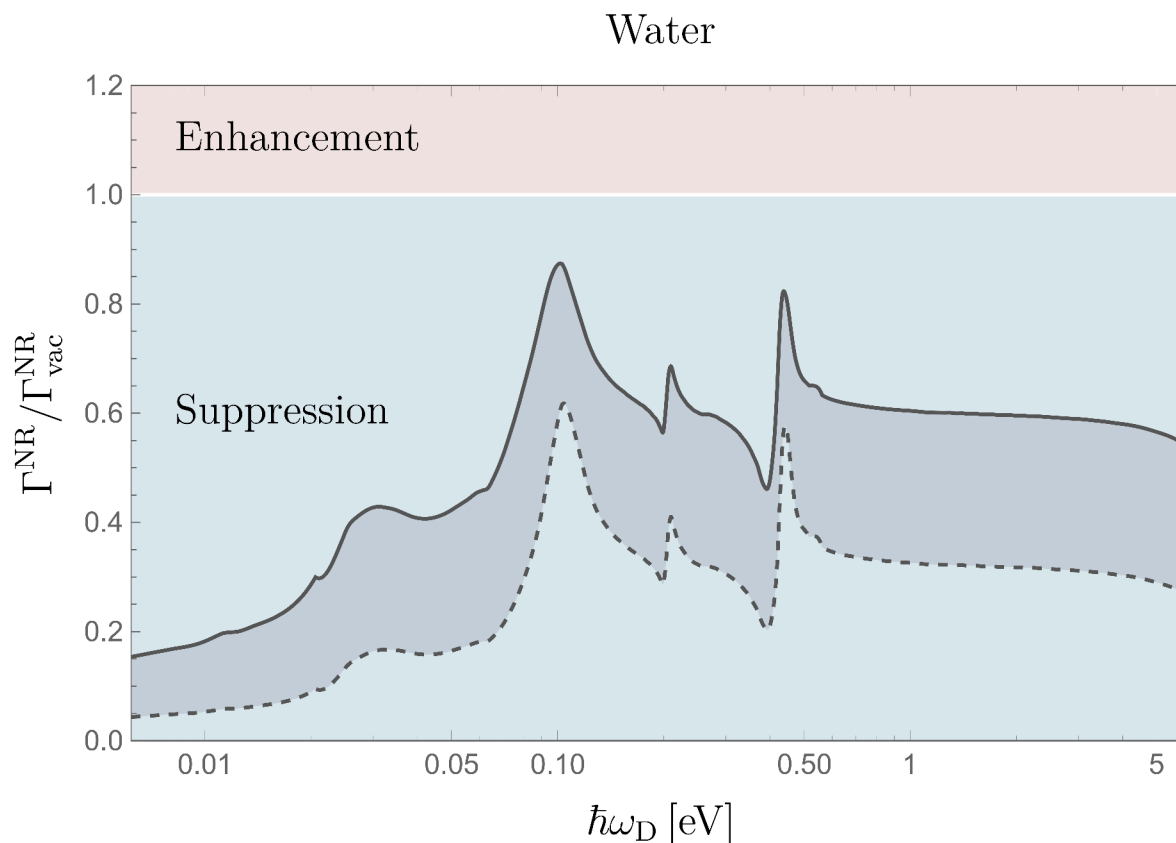
Retardation

$$\Gamma = \begin{cases} \sum_{k < n} \frac{3c^4 \Gamma_{n \rightarrow k}^D \sigma_A(\hbar\omega_{nk})}{4\omega_{nk}^4 r_{DA}^6} & \text{(nonretarded)} \\ \sum_{k < n} \frac{\Gamma_{n \rightarrow k}^D \sigma_A(\hbar\omega_{nk})}{4r_{DA}^2} & \text{(retarded)} \end{cases}$$



Bulk medium

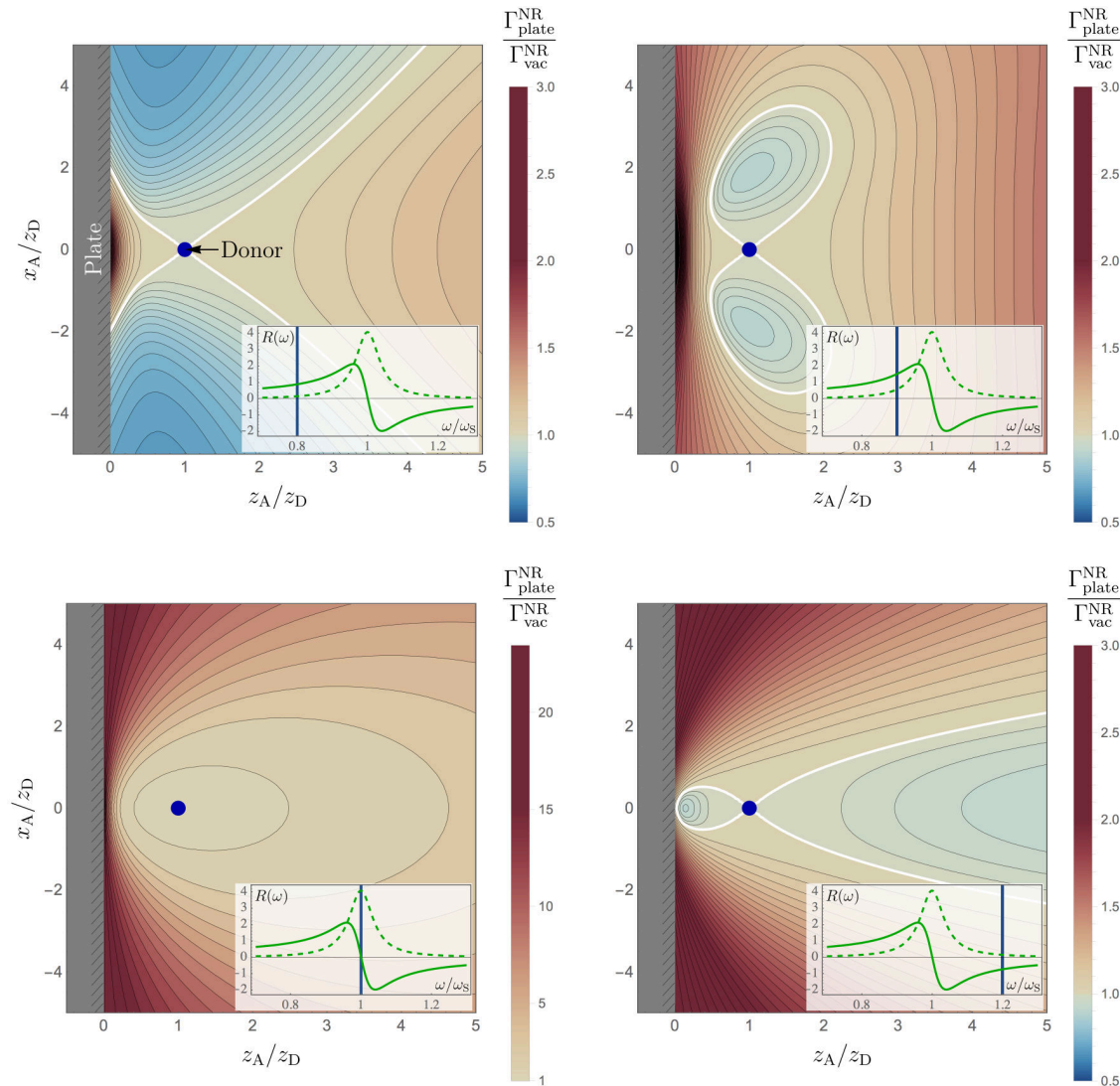
ICD rate in medium: $\Gamma = \frac{1}{|\epsilon|^2} \left| \frac{3\epsilon}{2\epsilon + 1} \right|^4 \Gamma^{(0)}$



Effect of medium: screening by polarisable medium

J. L. Hemmerich, R. Bennett and S.Y.B., Nature Commun. **9**, 2934 (2018)

Surface



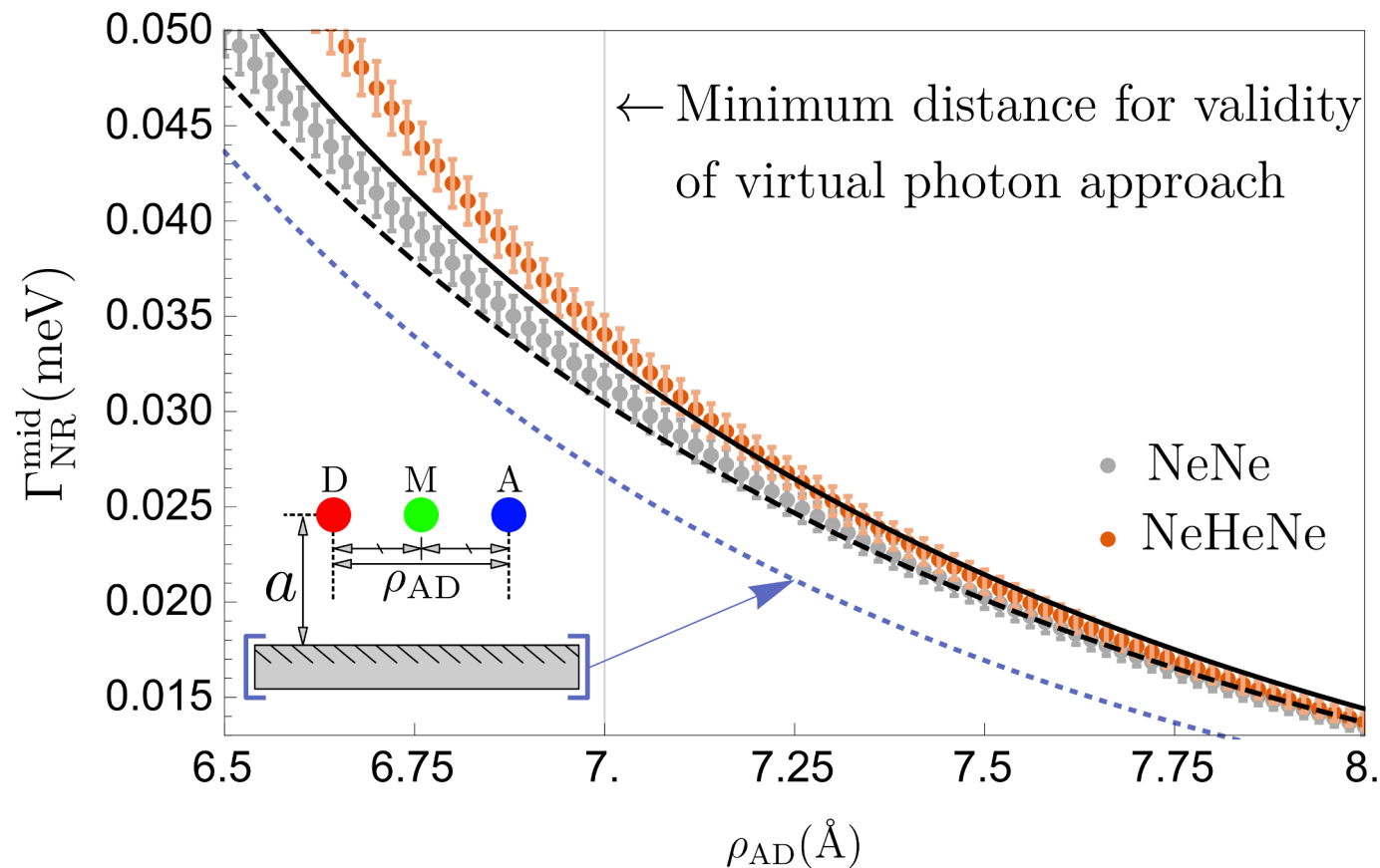
Effect of surface: position-dependent enhancement/suppression

J. L. Hemmerich, R. Bennett and S.Y.B., Nature Commun. **9**, 2934 (2018)

Mediator atom

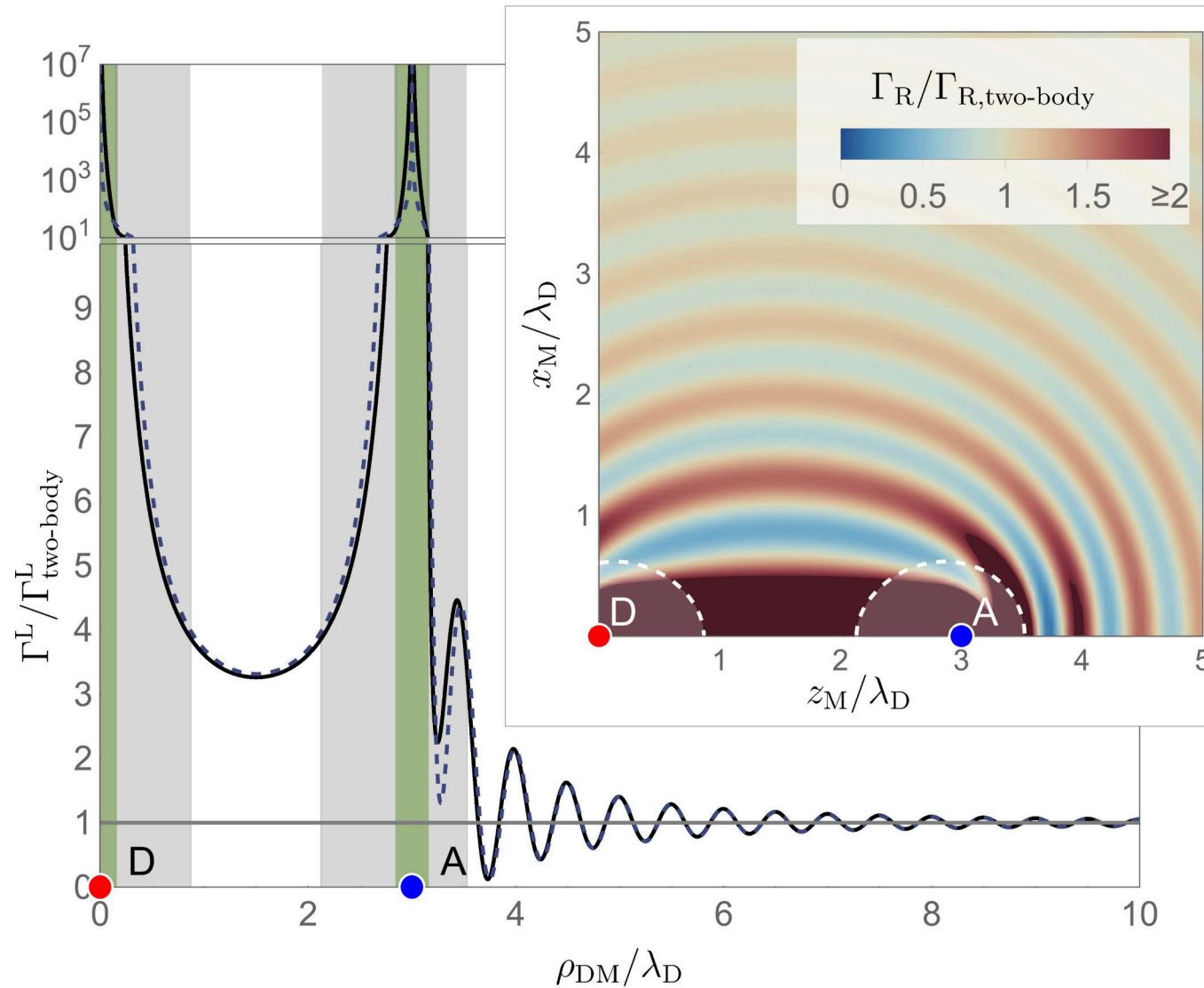
Passive mediator: polarisability α_M

$$\mathbf{G}(r, r') = \mathbf{G}^{(0)}(r, r') + \mu_0 \omega^2 \alpha_M \mathbf{G}^{(0)}(r, r_M) \cdot \mathbf{G}^{(0)}(r_M, r')$$



T. Miteva et al., PRL **119**, 083403 (2017);
 R. Bennett et al., PRL **122**, 153401 (2019)

Mediator atom



T. Miteva et al., PRL **119**, 083403 (2017);
R. Bennett et al., PRL **122**, 153401 (2019)

Acknowledgements



Postdocs: R. Bennett, J. Fiedler, F. Suzuki

PhD: S. Esfandiarpour, Y. Gorbachev,
J. Franz, S. Fuchs,
(P. Barcellona, J. Klatt)

MSc: S. Bang, F. Lindel, N. Strauß,
(F. Burger, J. Durnin, J. Hemmerich)

BSc: (V. Gebhart, T. Haug, M. Könné,
R. Oude Weernink, S. Rode)



Endress+Hauser

Conclusions

Macroscopic QED: atoms, photons, bodies

Atom–surface interactions: surface plasmons

⇒ non-additive laser-induced surface potential

⇒ enhanced quantum friction

Cavity QED: effective modes

⇒ position-dependent two-atom Rabi coupling

⇒ photon BEC as chiral sensor

Resonance energy transfer: impact of environments

⇒ solvent medium, surface, mediator

