Cooperative quantum optics with molecules

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MAX PLANCK INSTITUTE
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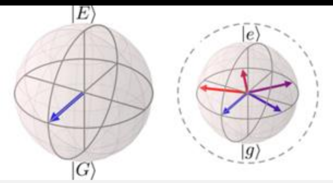
SPICE-Workshop on Quantum Spinoptics

-- 13.06.2023 --

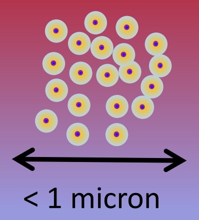


Genes Research Group
Cooperative Quantum Phenomena

Quantum at the collective level (from simple to complex quantum emitters)

1950s

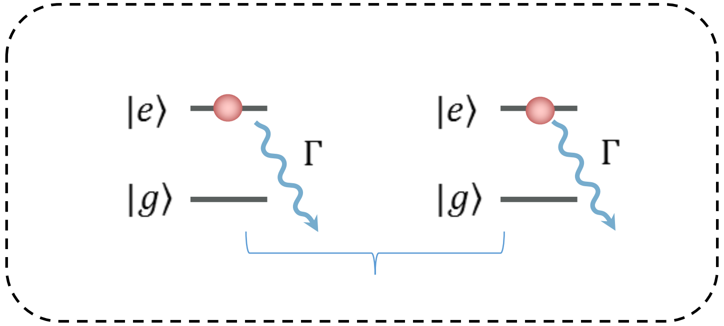
Dicke superradiance



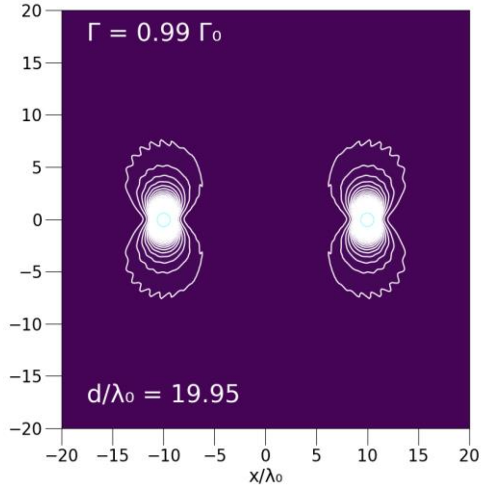
Enhanced collective emission

Two quantum emitters: superradiance and subradiance

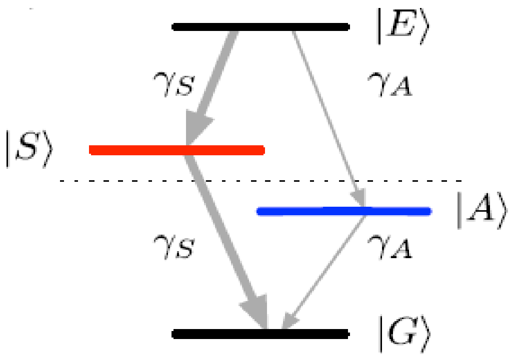
r<<λ



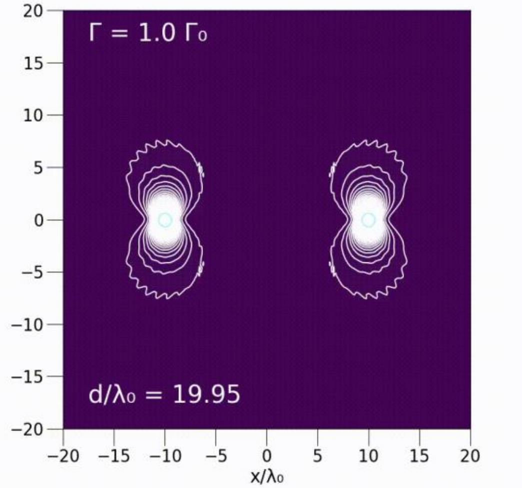
$|S\rangle = |eg\rangle + |ge\rangle$



Courtesy of Raphael Holzinger



$|A\rangle = |eg\rangle - |ge\rangle$



Robust states – quantum metrology, quantum information

State of the art

Cooperative Quantum Phenomena in Light-Matter Platforms

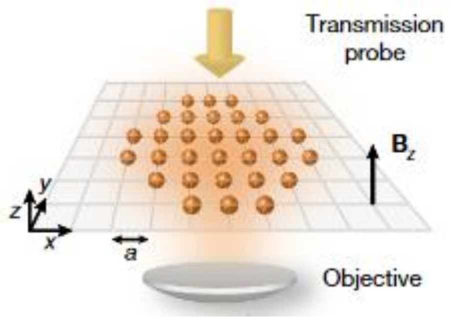
Michael Reitz[1,2] Christian Sommer[1,†] and Claudiu Genes[1,2,*]

PRX QUANTUM (TUTORIAL) 3, 010201 (2022) arxiv: 2107.02674

A subradiant optical mirror formed by a single structured atomic layer

https://doi.org/10.1038/s41586-020-2463-x
Received: 3 January 2020

Jun Rui[1,2]✉, David Wei[1,2], Antonio Rubio-Abadal[1,2], Simon Hollerith[1,2], Johannes Zeiher[3], Dan M. Stamper-Kurn[3], Christian Gross[1,2,4] & Immanuel Bloch[1,2,5]



- Strong and narrow cooperative subradiant response
- Only a few hundred atoms (extremely small mass)
- Efficient optical metamaterial engineering
- Applications in low-mass hybrid nano-optomechanics

Optical metasurfaces

State of the art

Cooperative Quantum Phenomena in Light-Matter Platforms

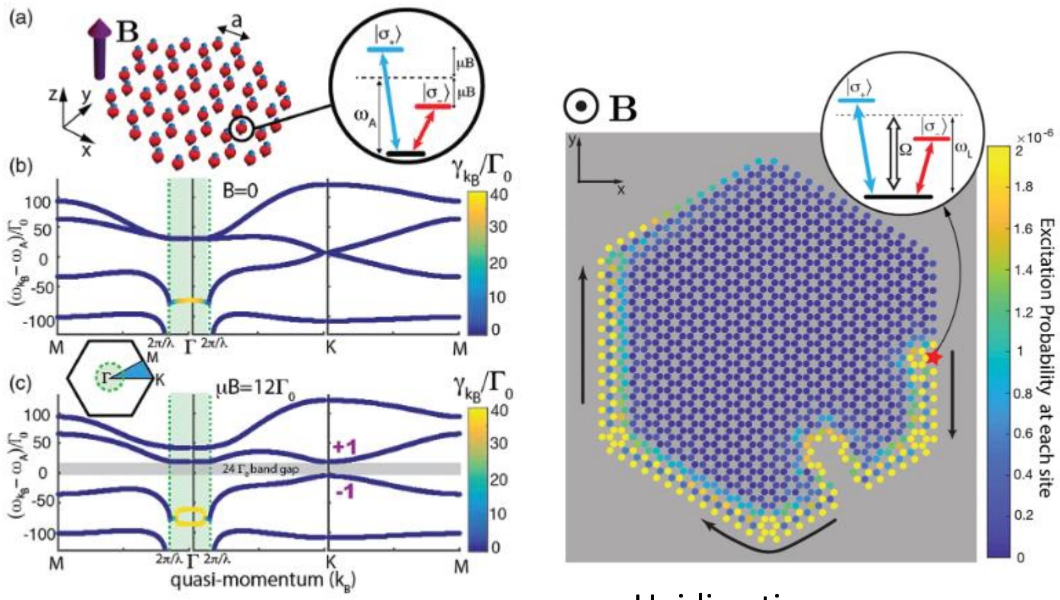
Michael Reitz[1,2] Christian Sommer[1,†] and Claudiu Genes[1,2,*]

PRX QUANTUM (TUTORIAL) 3, 010201 (2022) arxiv: 2107.02674

PRL 119, 023603 (2017) PHYSICAL REVIEW LETTERS week ending 14 JULY 2017

Topological Quantum Optics in Two-Dimensional Atomic Arrays

J. Perczel,[1,2] J. Borregaard,[2] D. E. Chang,[3] H. Pichler,[2,4] S. F. Yelin,[2,5] P. Zoller,[6,7] and M. D. Lukin[2]



- Unidirection energy transport around large defects

Platforms for excitation transport

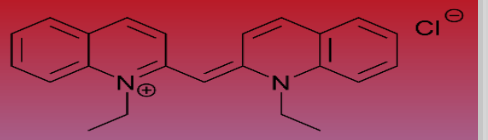
Quantum at the collective level (from simple to complex quantum emitters)

1930s

1950s

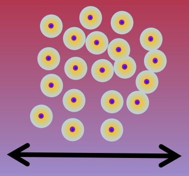
Molecular aggregates

PIC chloride



Supramolecular structures

Dicke superradiance



< 1 micron

Enhanced collective emission

E. E. Jelley 1936
G. Scheibe 1937

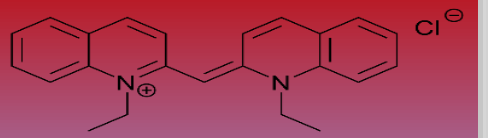
- Type of dye with increased absorption coefficient after aggregation (aided by solvent)

Quantum at the collective level (from simple to complex quantum emitters)

1930s | 1950s | 1950s

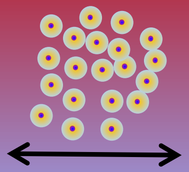
Molecular aggregates

PIC chloride



Supramolecular structures

Dicke superradiance

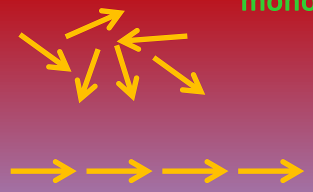


< 1 micron

Enhanced collective emission

Kasha's theory

monomers



aggregates

- Collective dipole absorption
- Inhibition of fluorescence in H-aggregates

E. E. Jelley 1936
G. Scheibe 1937

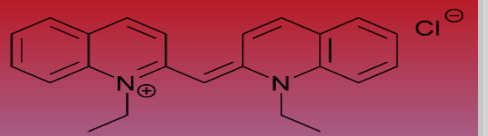
- Type of dye with increased absorption coefficient after aggregation (aided by solvent)

Quantum at the collective level (from simple to complex quantum emitters)

1930s	1950s	1950s	2000s -2020s
Molecular aggregates	**Dicke superradiance**	**Kasha's theory**	**Cavity quantum materials**

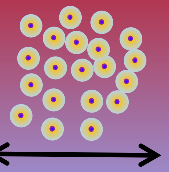
Molecular aggregates

PIC chloride



Supramolecular structures

Dicke superradiance

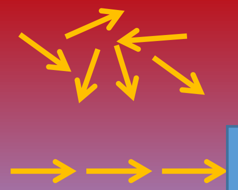


< 1 micron

Enhanced collective emission

Kasha's theory

monomers

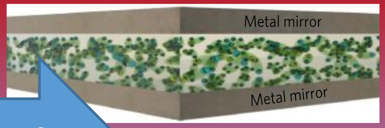


aggregates

- Collective dipole absorption
- Inhibition of fluorescence in H-aggregates

Extremely large dipole moments

Cavity quantum materials



ry-Perot / planar cavities
Sanvitto & Kéna-Cohen, Nat. Mater. **15**, 1061 (2016)

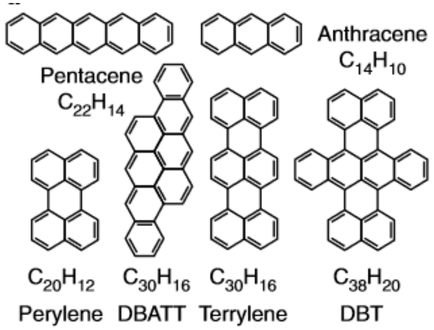
Strong **collective** light-matter interactions

Why molecules

Single organic molecules for photonic quantum technologies

NATURE MATERIALS | VOL 20 | DECEMBER 2021 | 1615–1628 |

C. Toninelli[1,2], I. Gerhardt[3], A. S. Clark[4], A. Reserbat-Plantey[5], S. Götzinger[6,7], Z. Ristanović[8], M. Colautti[1,2], P. Lombardi[1,2], K. D. Major[4], I. Deperasińska[9], W. H. Pernice[10], F. H. L. Koppens[5,11], B. Kozankiewicz[9], A. Gourdon[12], V. Sandoghdar[6,7] and M. Orrit[8]



Polyaromatic hydrocarbons (PAH)

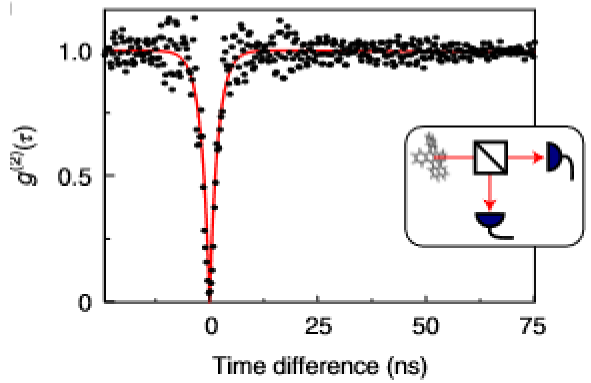
Advantages

- Good isolation in solid state host matrices
- Flexibility in synthesis – wide pallete of emission wavelengths
- Optimized interaction with light

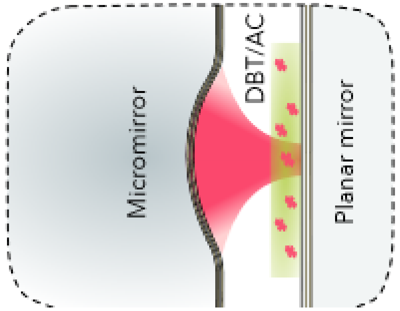
Promises

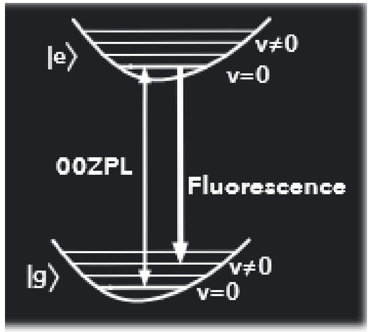
- Single photon sources
- Nonlinear elements with competitive performance in terms of coherence, scalability and compatibility with diverse integrated platforms
- Transducers – promise of single quanta resolution in the sensing of charges and motion

Photon antibunching



Qubit - closed two level system





Wang, D. et al. Turning a molecule into a coherent two-level quantum system. Nat. Phys. 15, 483–489 (2019)

Why molecules

Use light (*vacuum field*) to modify material properties?

Confine light in space to get strong few-photon interaction with interesting materials

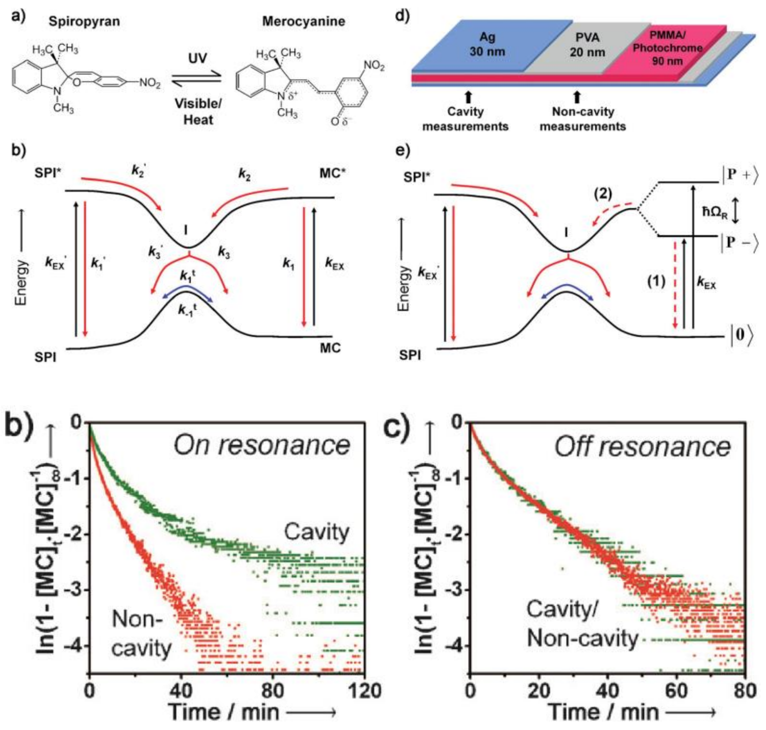
Many molecules / macroscopic

Promises

- ”Easy” to reach collective strong coupling
- Modifications of electronic properties
- Tunable chemistry
- Could integrate in devices

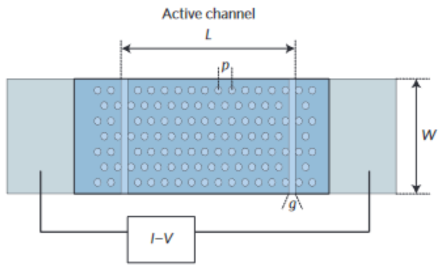
Modify photochemical reaction rates

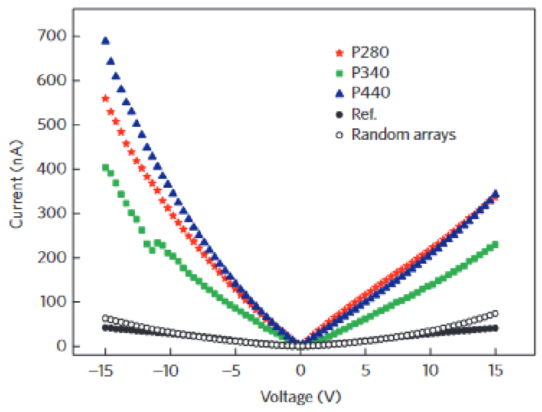
J. A. Hutchison *et al.*, Angew. Chemie **124**, 1624 (2012)



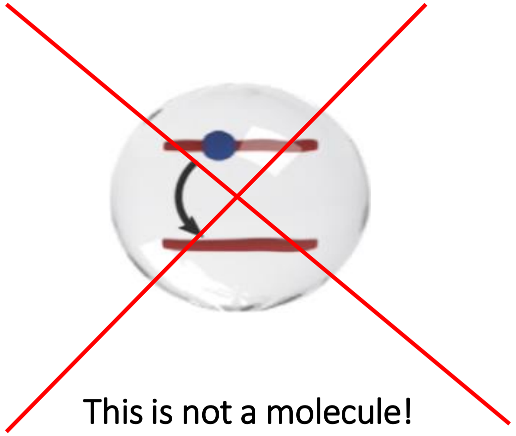
Conductivity in organic semiconductors hybridized with the vacuum field

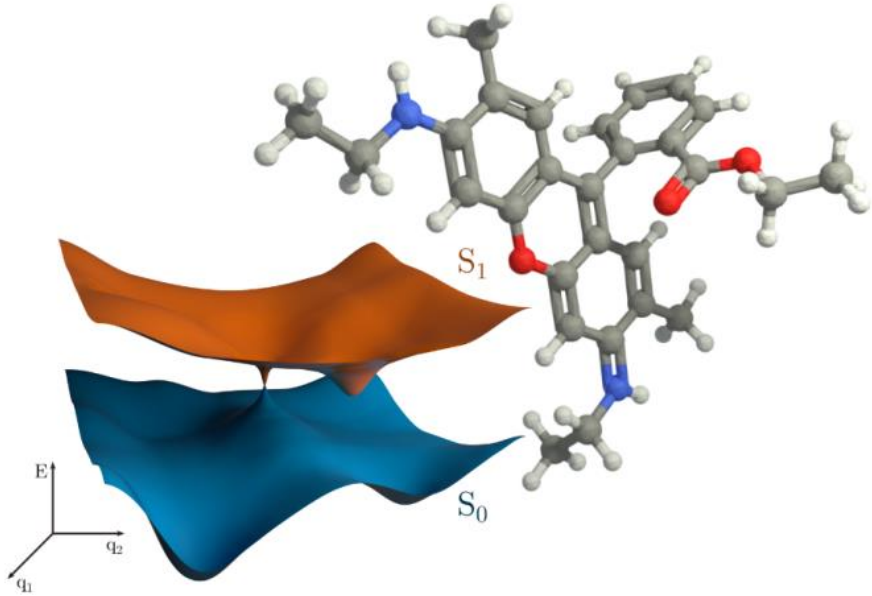
E. Orgiu et al., Angew. Nat Mat **14**, 1123 (2015)





The complex nature of molecules

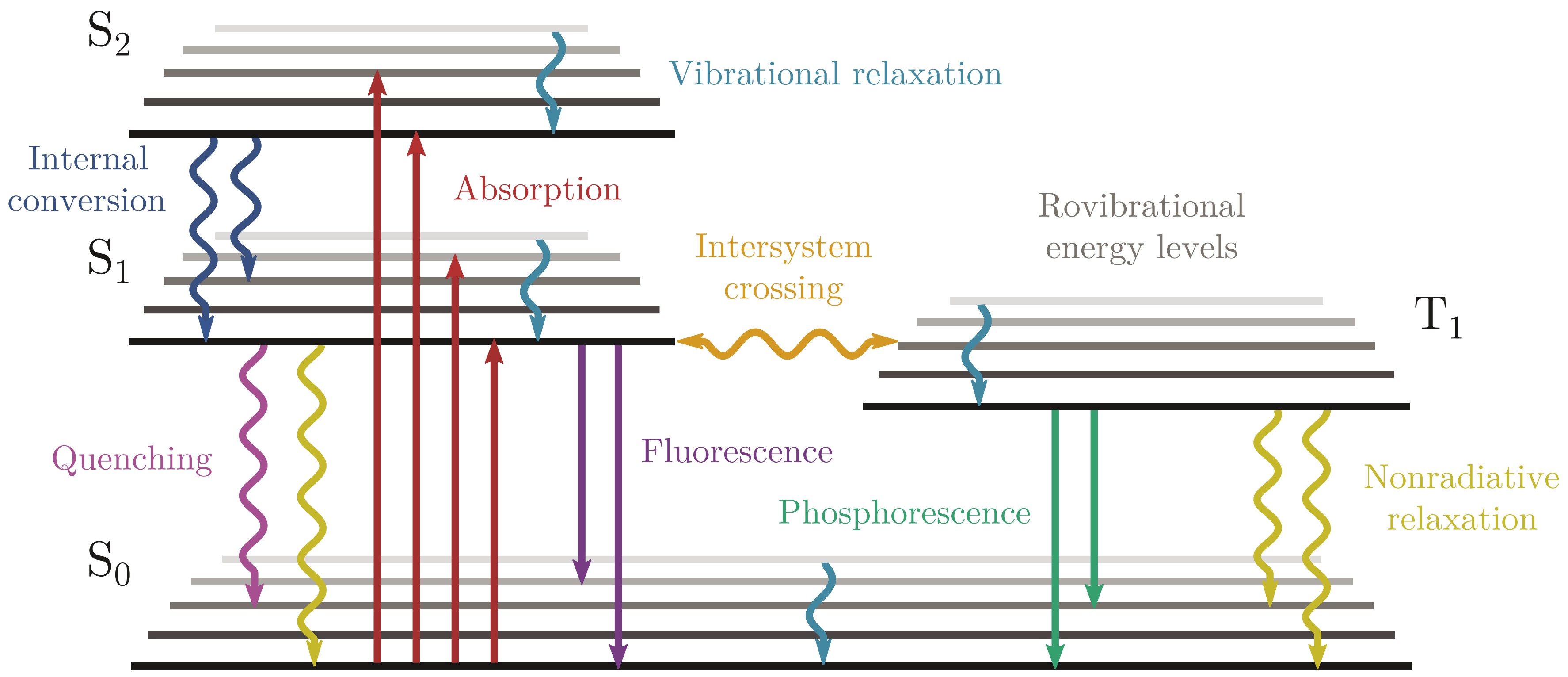




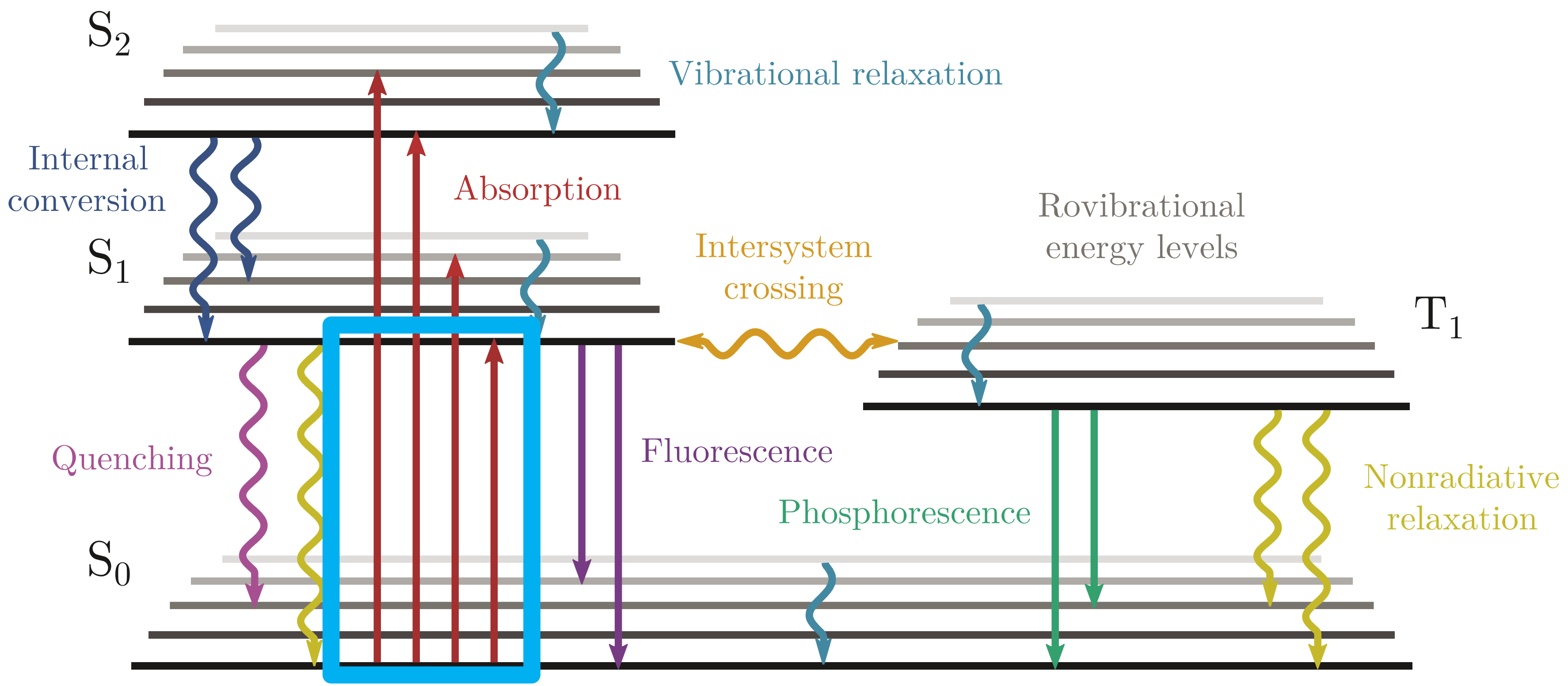
Fundamental aspects

- Photon-electron coupling strongly perturbed by additional vibrational degrees of freedom
- Radiative emission can compete with non-radiative pathways of relaxation

The complex nature of molecules



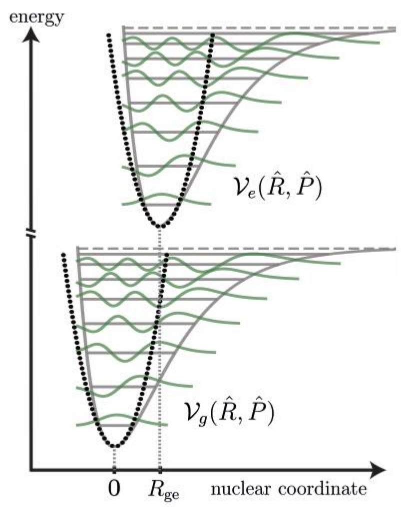
The complex nature of molecules

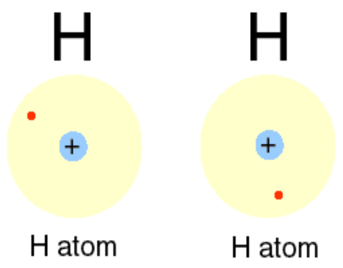


Coupling at optical frequencies

- Experimentally: photo-physics, (photo) chemistry, charge/exciton transport, etc.
- Theory: **role of vibrations**, Tavis-Cummings-Holstein model, polariton cross-talk, etc...

Quantum optics approach to molecules: a simple model

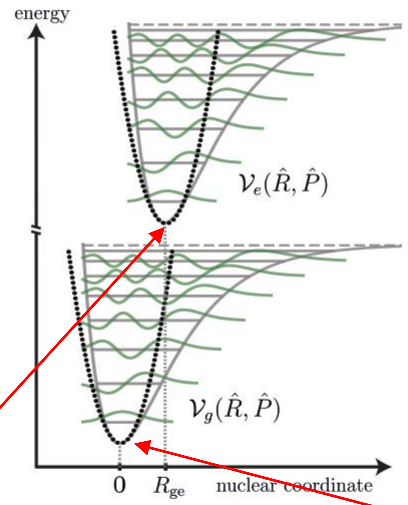


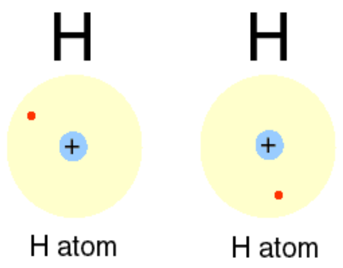


Minimal model

- Expansion of the molecular potential landscapes along the nuclear coordinate
- Harmonic approximation
- Difference between minima gives rise to electron-vibron coupling

Quantum optics approach to molecules: a simple model





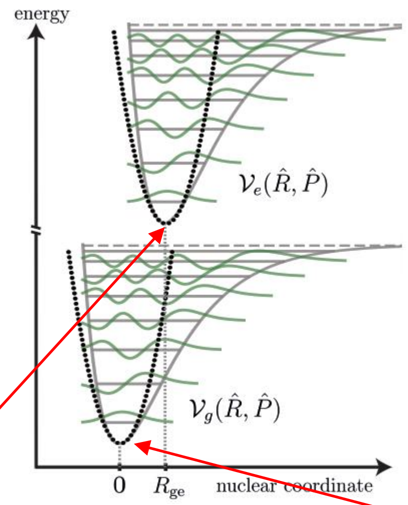
$$H = \left(\omega_e + \frac{p^2}{2\mu} + \frac{1}{2}\mu\nu^2\,(R - R_e)^2\right)\sigma^\dagger\sigma + \left(\frac{p^2}{2\mu} + \frac{1}{2}\mu\nu^2\,(R - R_g)^2\right)\sigma\sigma^\dagger$$

Electronic operators (spin algebra)

Minimal model

- Expansion of the molecular potential landscapes along the nuclear coordinate
- Harmonic approximation
- Difference between minima gives rise to electron-vibron coupling

Quantum optics approach to molecules



$$H = \left(\omega_e + \frac{p^2}{2\mu} + \frac{1}{2}\mu\nu^2\left(R - R_e\right)^2\right)\sigma^\dagger\sigma + \left(\frac{p^2}{2\mu} + \frac{1}{2}\mu\nu^2\left(R - R_g\right)^2\right)\sigma\sigma^\dagger$$

Electronic operators (spin algebra)

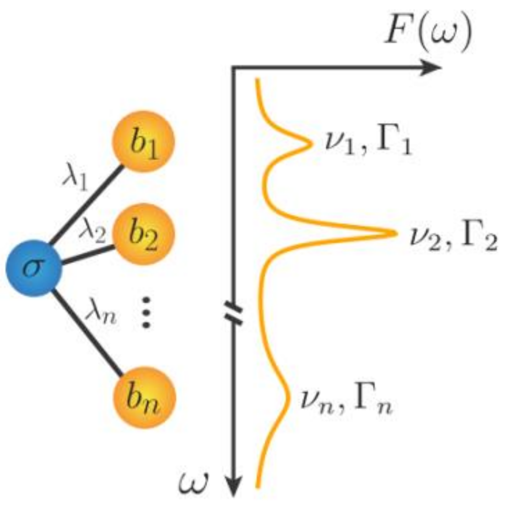
Holstein Hamiltonian

$$H = \nu b^\dagger b + (\omega_e + \lambda^2\nu)\sigma^\dagger\sigma - \lambda\nu(b^\dagger + b)\sigma^\dagger\sigma$$

Huang-Rhys factor

Bosonic operators (for vibrations)

Quantum Langevin equations



$$\frac{db_k(t)}{dt} = -(\Gamma_k + i\nu_k)b_k(t) + \sqrt{2\Gamma_k}b_{in}^k(t),$$
$$\frac{d\tilde{\sigma}(t)}{dt} = -[\gamma - i(\omega_\ell - \omega_e)]\,\tilde{\sigma}(t) + \sqrt{2\gamma}\mathcal{D}^\dagger(t)\Sigma_{in}(t)$$

Polaron operator

$$\tilde{\sigma}(t) = \sigma(t)\mathcal{D}^\dagger(t)$$
$$\mathcal{D}^\dagger = \mathcal{D}_1^\dagger ... \mathcal{D}_n^\dagger$$

Thermal reservoir

$$\langle b_{in}^\dagger(t)b_{in}(t')\rangle = \Gamma n_T \delta(t-t')$$
$$\langle b_{in}(t)b_{in}^\dagger(t')\rangle = \Gamma(n_T+1)\delta(t-t')$$
$$n_T = [\exp(\hbar\nu/(k_B T)) - 1]^{-1}$$

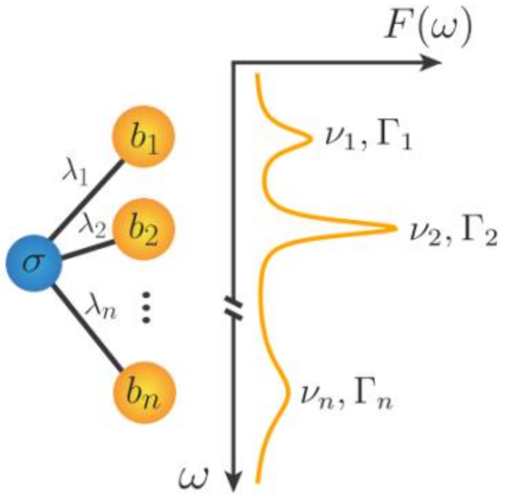
Input non-zero noise

$$\Sigma_{in}(t) = \eta_\ell/\sqrt{2\gamma} + \sigma_{in}(t)$$

Solution

$$\sigma(t) = \sigma(0)\mathcal{D}(t)\mathcal{D}^\dagger(0)e^{-[\gamma-i(\omega_\ell-\omega_e)]t} + \sqrt{2\gamma}\int_0^t dt' e^{-[\gamma-i(\omega_\ell-\omega_e)](t-t')}\Sigma_{in}(t')\mathcal{D}(t)\mathcal{D}^\dagger(t')$$

Quantum Langevin equations



$$\frac{db_{\mathrm{k}}(t)}{dt} = -(\Gamma_{\mathrm{k}} + i\nu_{\mathrm{k}})b_{\mathrm{k}}(t) + \sqrt{2\Gamma_{\mathrm{k}}}b_{\mathrm{in}}^{\mathrm{k}}(t),$$

$$\frac{d\tilde{\sigma}(t)}{dt} = -\left[\gamma - i(\omega_\ell - \omega_{\mathrm{e}})\right]\tilde{\sigma}(t) + \sqrt{2\gamma}\mathcal{D}^\dagger(t)\Sigma_{\mathrm{in}}(t)$$

Polaron operator

$$\tilde{\sigma}(t) = \sigma(t)\mathcal{D}^\dagger(t)$$
$$\mathcal{D}^\dagger = \mathcal{D}_1^\dagger \ldots \mathcal{D}_n^\dagger$$

Thermal reservoir

From here on:

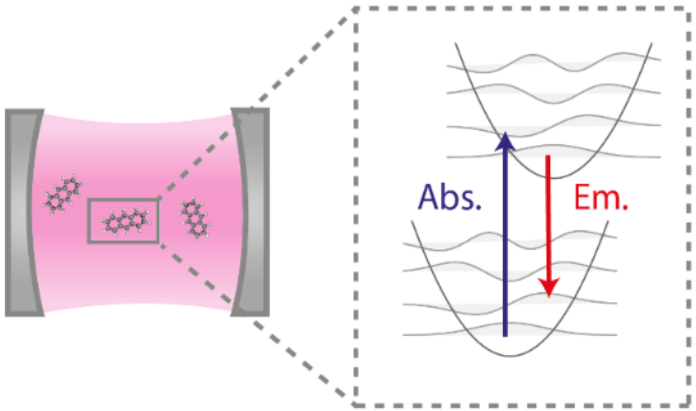
- Follow standard quantum optics techniques
- Two-time correlations lead to absorption and emission spectra
- Easy to add dynamics inside optical cavities

Solution

$$\sigma(t) = \sigma(0)\mathcal{D}(t)\mathcal{D}^\dagger(0)e^{-[\gamma-i(\omega_\ell-\omega_{\mathrm{e}})]t}$$

Results

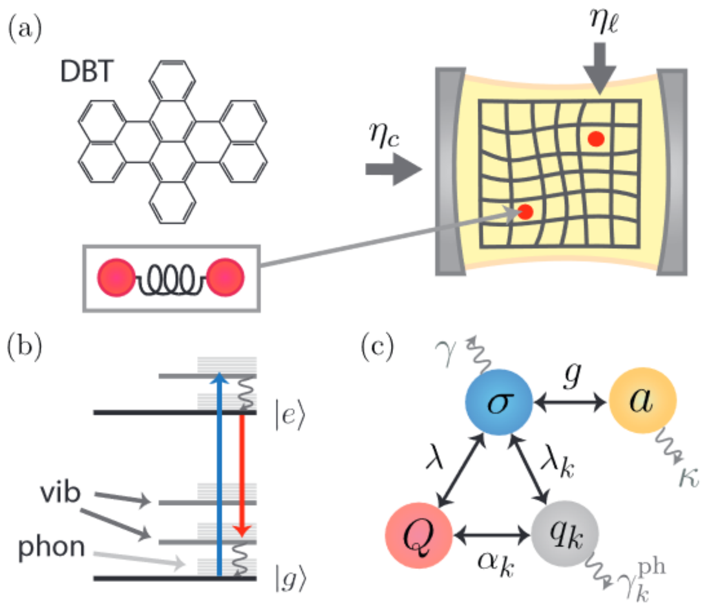
Cavity QED with single or few molecules



Results

- Analytical approach to electron-photon-vibron
- Polariton cross-talk rates
- Turning a molecule into a single closed qubit
- Cavity modified Förster resonance energy transfer

M. Reitz, C. Sommer and C. Genes, **Phys. Rev. Lett. 122, 203602 (2019)**
Langevin approach to quantum optics with molecules

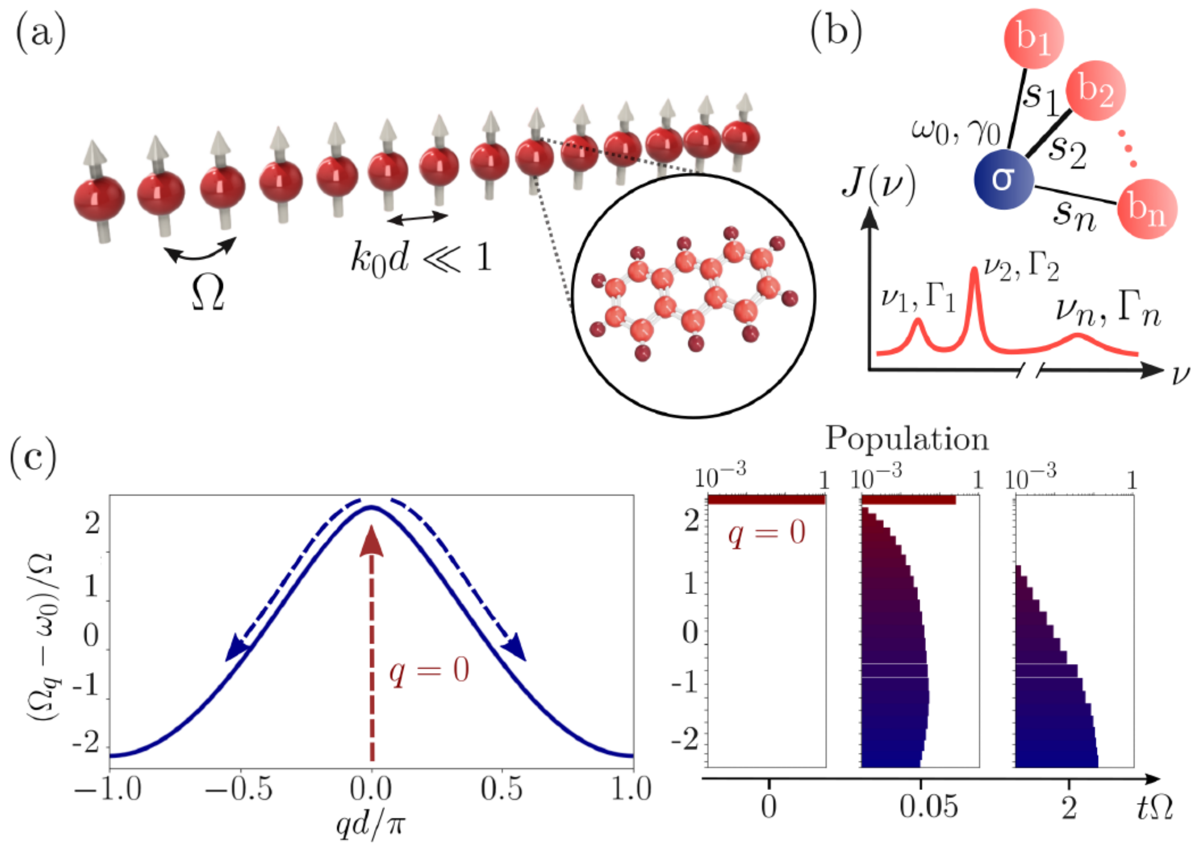


Results

- Analytical approach to electron-photon-vibron-phonon interactions
- Vibrational collective decoupling

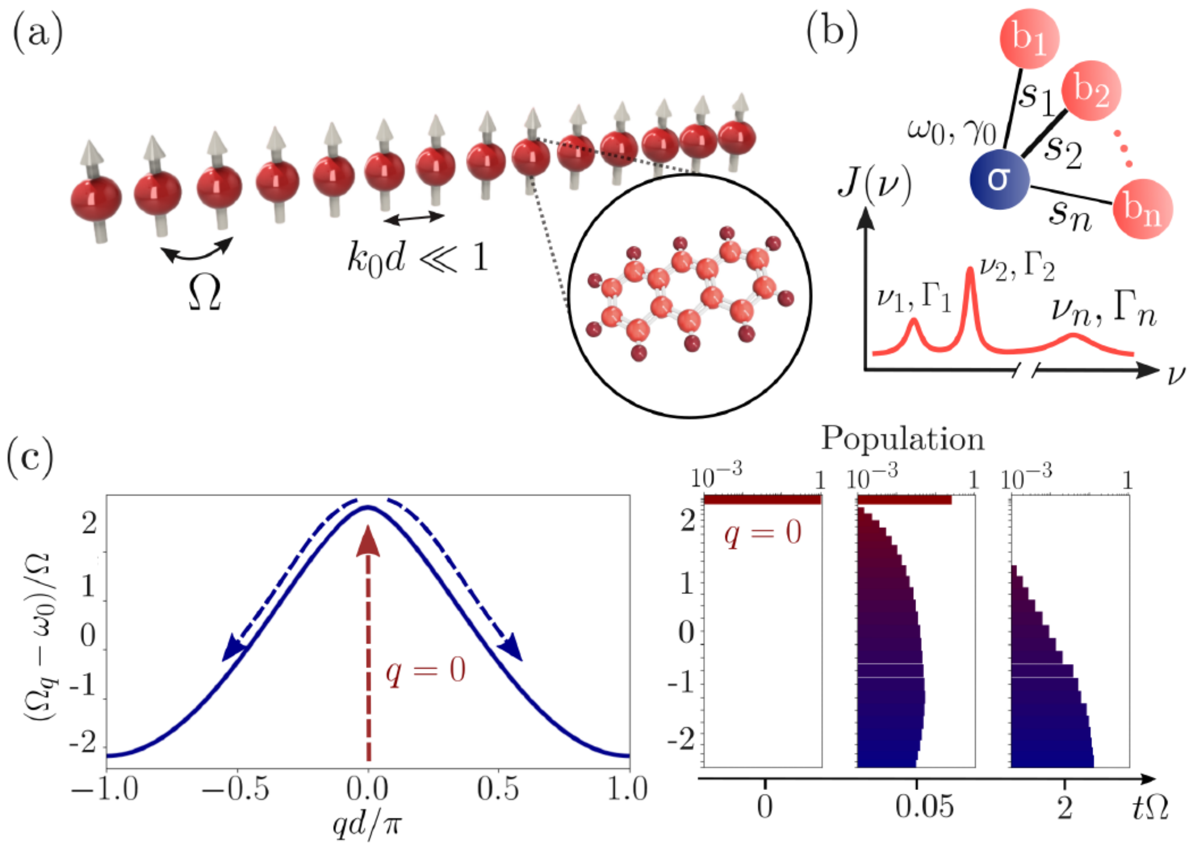
M. Reitz, C. Sommer, B. Gurlek, V. Sandoghdar, D. Martin-Cano and C. Genes,
Phys. Rev. Research 2, 033270 (2020)
Molecule-photon interactions in phononic environments

Molecular aggregates



R. Holzinger, N. D. Bassler, H. Ritsch and C. Genes, **arxiv:2304.10236 (2023),** *Scaling law for Kasha's rule in photoexcited subwavelength molecular aggregates*

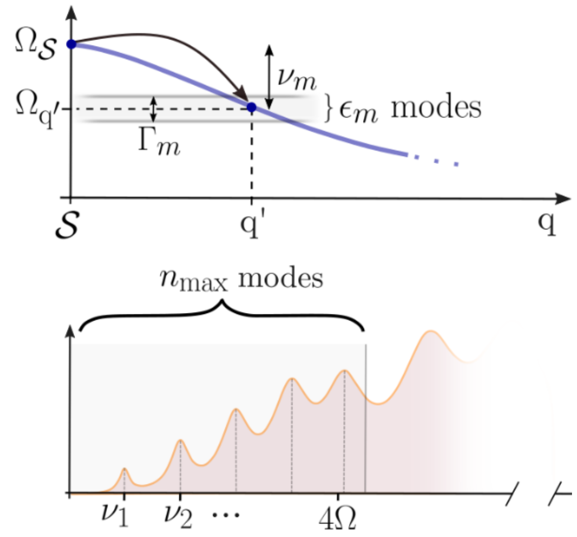
Molecular aggregates



Rate equations

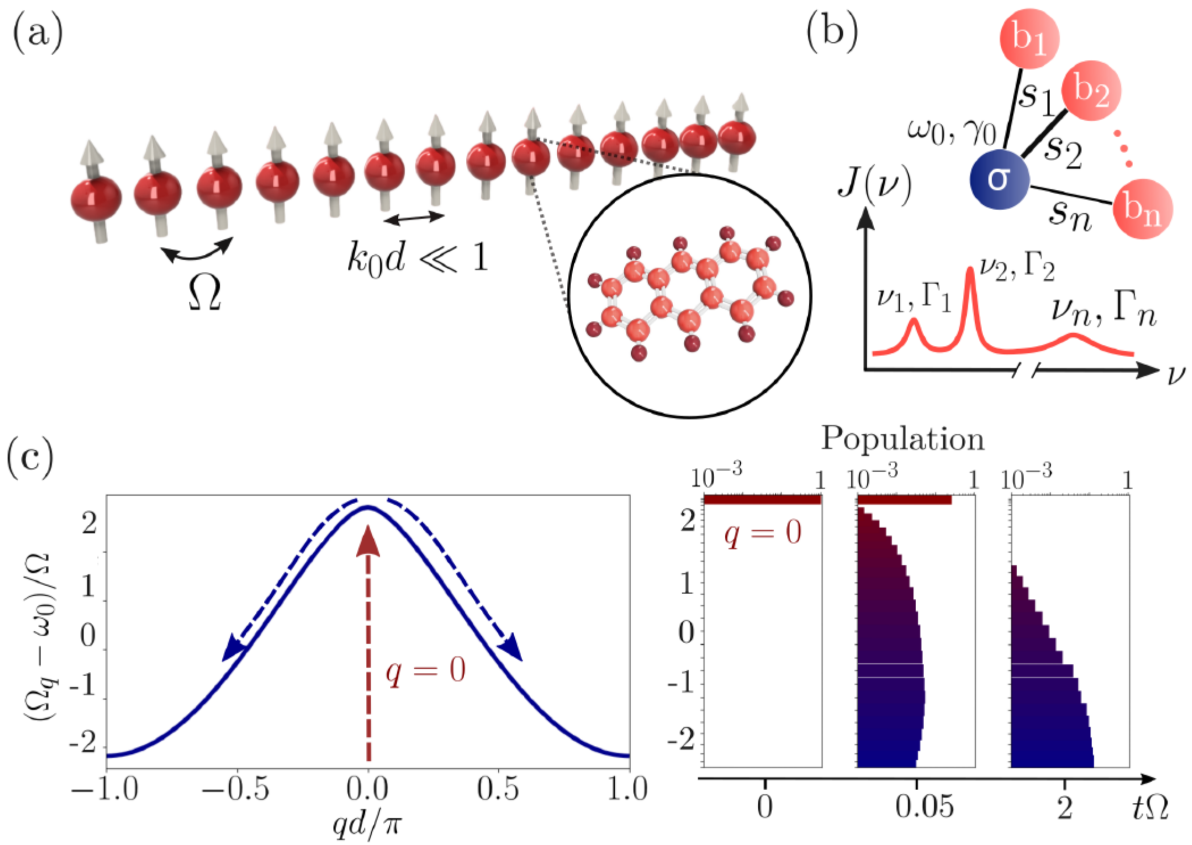
$$\dot{p}_{\mathcal{S}} = -(\gamma_{\mathcal{S}} + \kappa_{\mathcal{S}})p_{\mathcal{S}} + \sum_{q \neq 0} \kappa_{q \to \mathcal{S}} p_q,$$

$$\dot{p}_q = -\kappa_q p_q + \sum_{q' \neq q} \kappa_{q' \to q} p_{q'}.$$



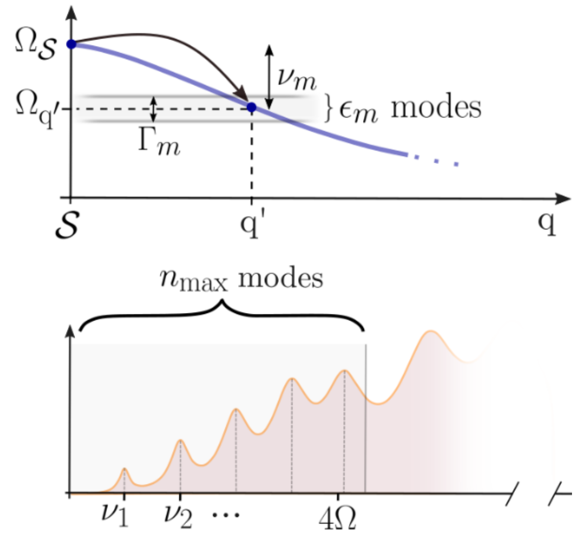
R. Holzinger, N. D. Bassler, H. Ritsch and C. Genes, **arxiv:2304.10236 (2023),** *Scaling law for Kasha's rule in photoexcited subwavelength molecular aggregates*

Molecular aggregates



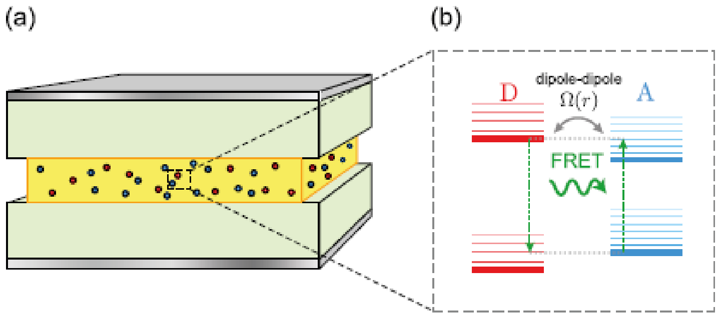
Scaling

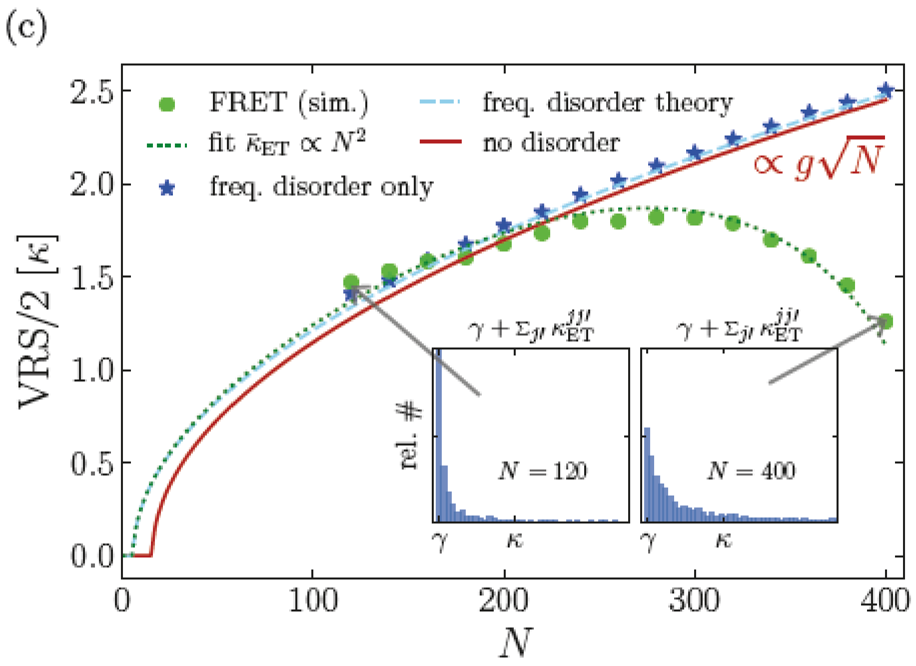
$$\kappa_{\mathcal{S}} \approx \frac{4s\Omega}{3} \frac{(n_{\max}+1)(2n_{\max}+1)}{n_{\max}}$$



R. Holzinger, N. D. Bassler, H. Ritsch and C. Genes, **arxiv:2304.10236 (2023),** *Scaling law for Kasha's rule in photoexcited subwavelength molecular aggregates*

Cavity QED with mescoscopic ensembles





Results

- Disorder provides loss of polaritons
- Disorder plus vibrations can reduce the Vacuum Rabi Splitting

C. Sommer, M. Reitz, F. Mineo and C. Genes, **Phys. Rev. Research 3, 033141 (2021)**
Molecular polaritonics in dense mesoscopic disordered ensembles

Thank you!