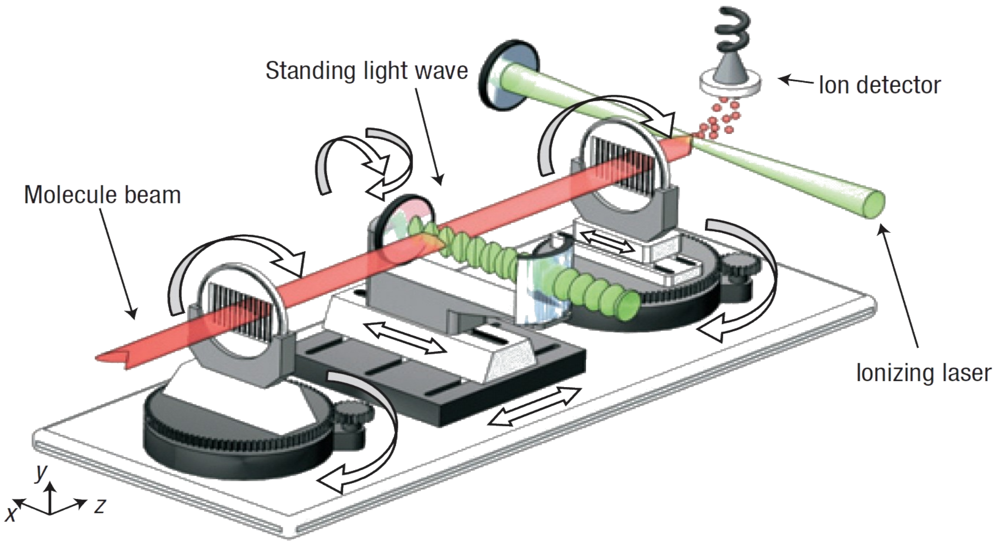
Klaus Hornberger

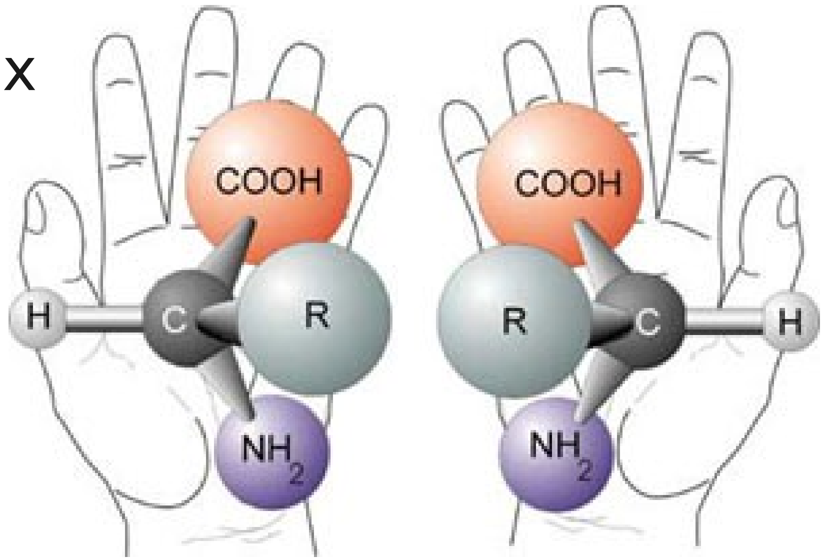
Decoherence phenomena in molecular systems

Overview

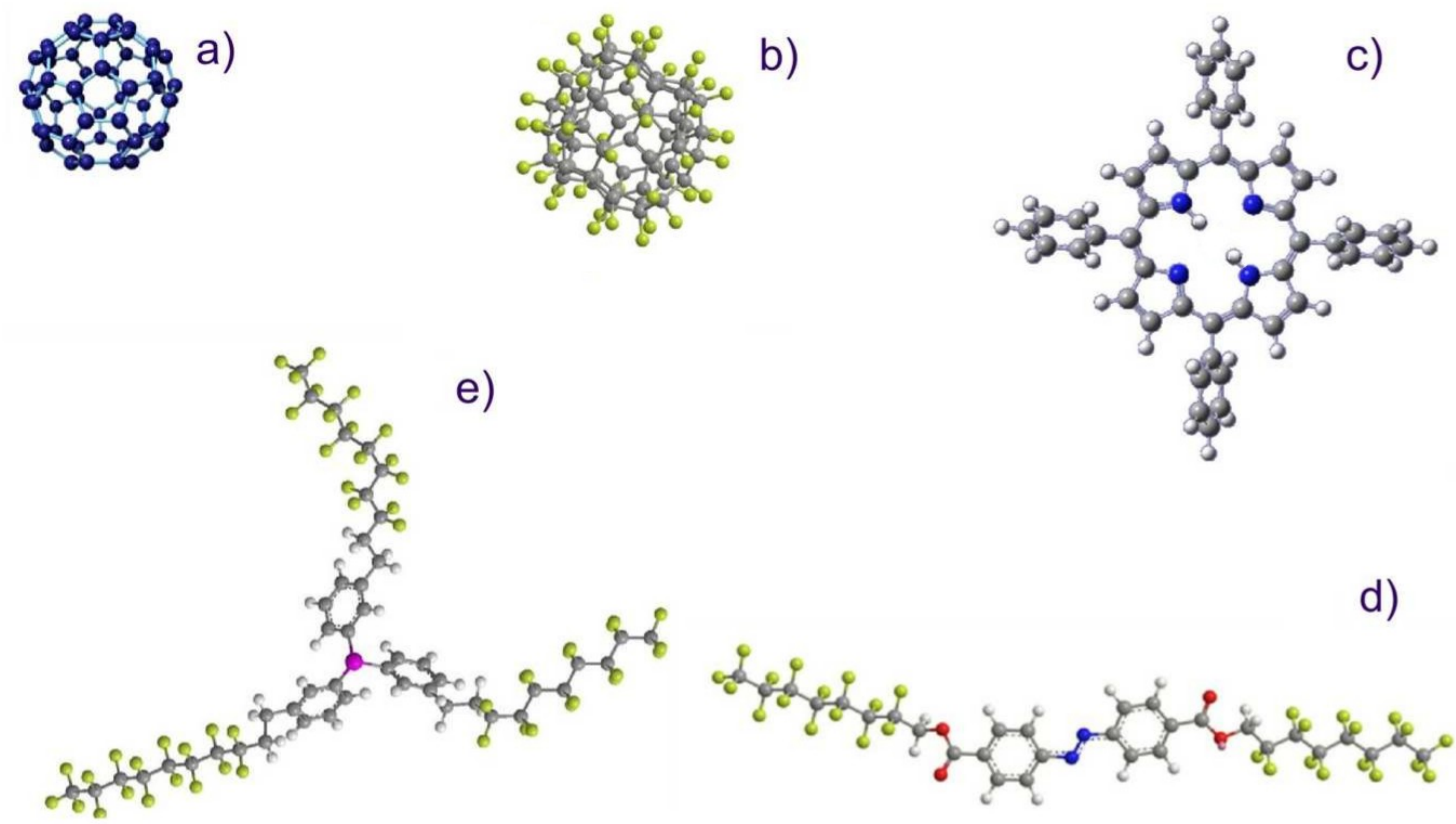
1. Localization of molecular matter waves



2. Hund's paradox of molecular chirality



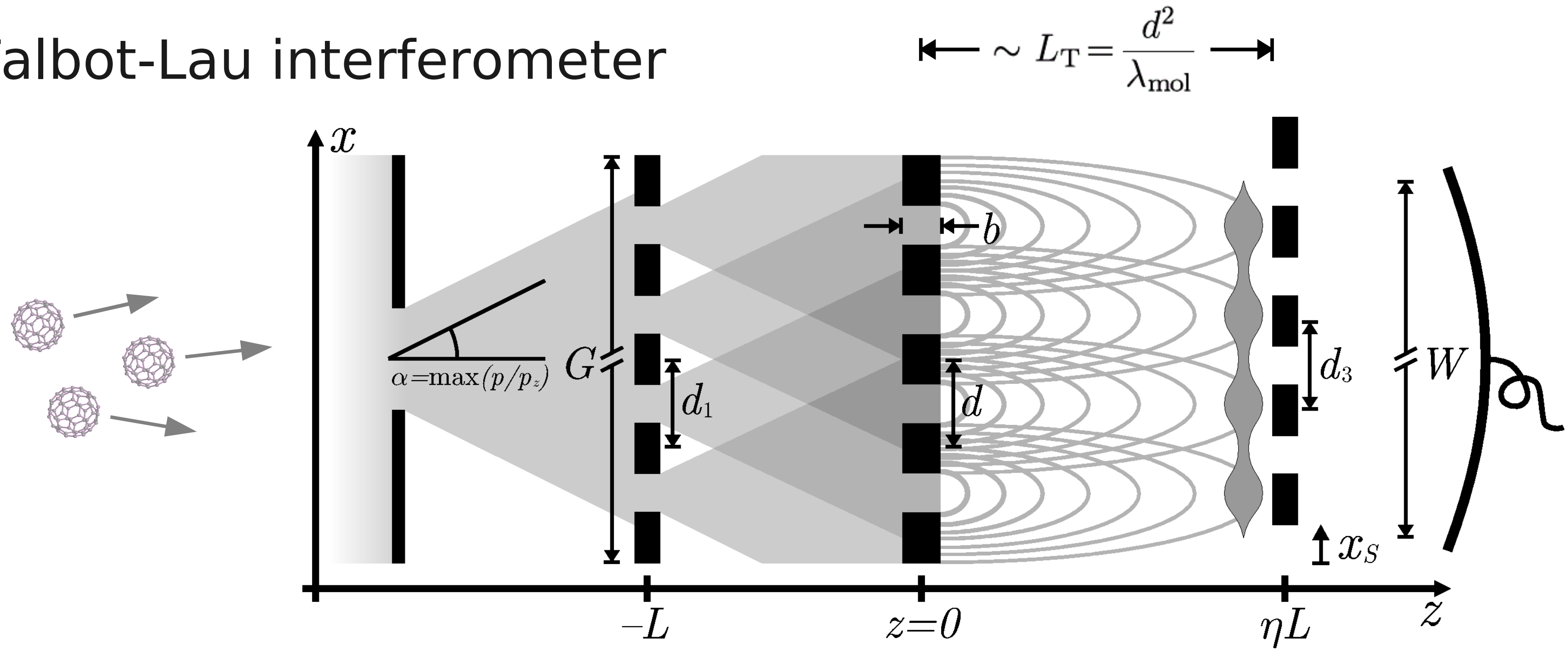
A gallery of molecules...



... whose wave nature has been observed (University of Vienna)

Molecular near-field diffraction

Talbot-Lau interferometer

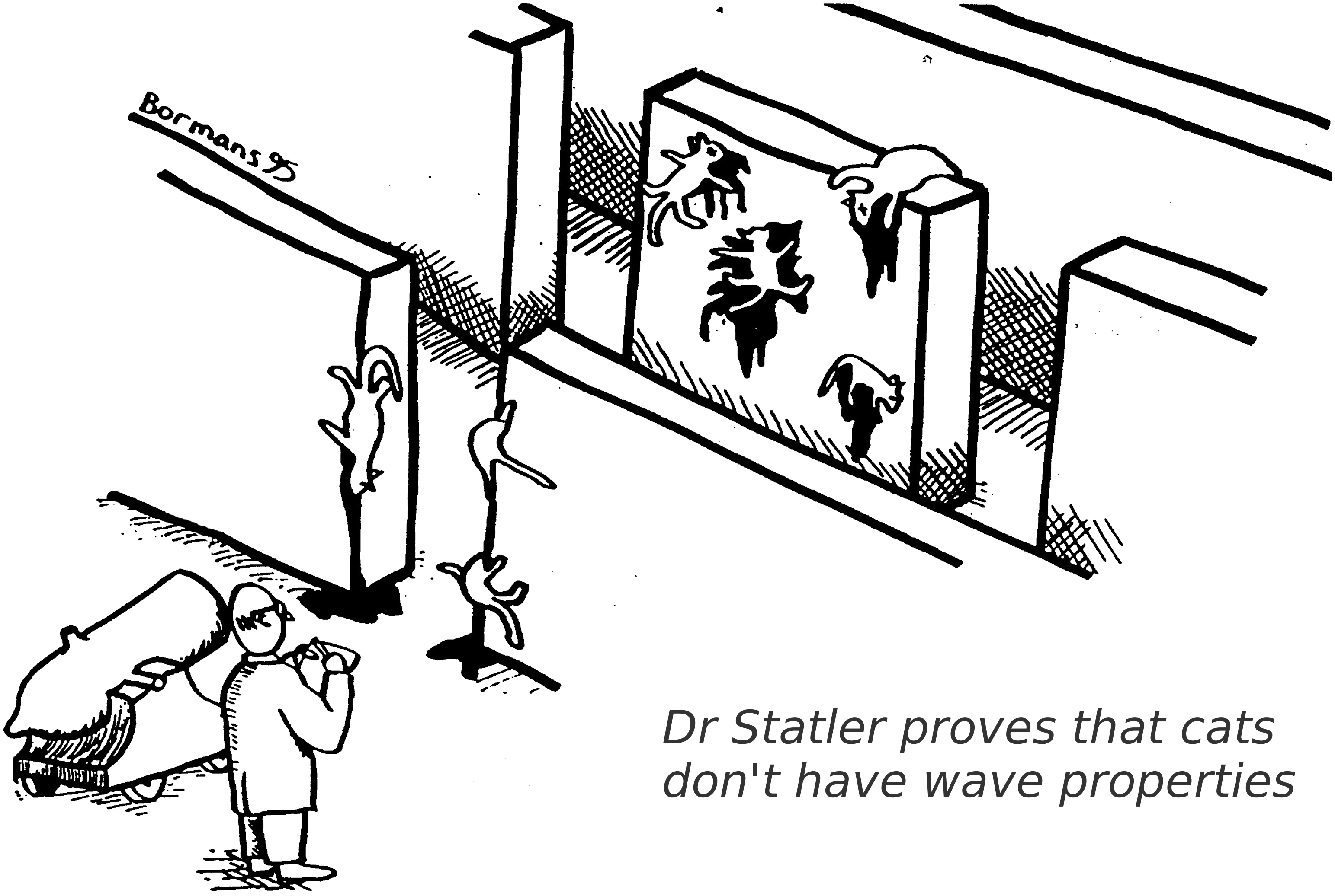


Phase space description allows for

- Comparison with classical formulation
- Description of decoherence effects
- Accounting for grating dispersion forces

Decoherence

...explains the gradual emergence of classical behavior due to unavoidable environmental interactions



Dr Statler proves that cats don't have wave properties

Localization in position

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle \boldsymbol{X}\,|\tilde{\rho}\,|\boldsymbol{X}'\rangle \;=\; -F(\boldsymbol{X}-\boldsymbol{X}')\,\langle \boldsymbol{X}\,|\tilde{\rho}\,|\boldsymbol{X}'\rangle$$

by endogenous heat radiation

$$F(\boldsymbol{x}) \;=\; \int_0^\infty \mathrm{d}\lambda\, R_\lambda(\lambda,T)\left(1-\mathrm{sinc}\left[2\pi\,\frac{|\boldsymbol{x}|}{\lambda}\right]\right)$$

spectral emission rate

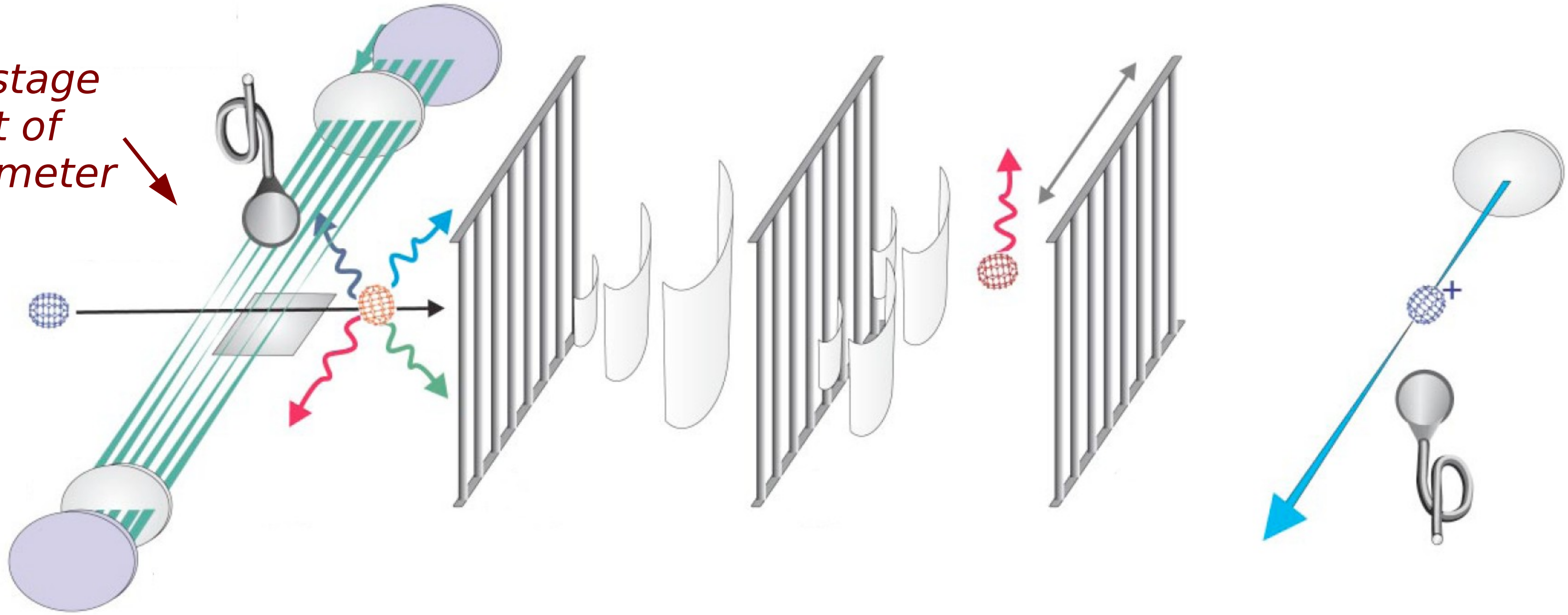
by gas collisions

$$F(\boldsymbol{x}) \;=\; n_{\mathrm{gas}}\left\langle v_{\mathrm{g}}\int \mathrm{d}\Omega\,\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\left(\cos\theta;\frac{m_{\mathrm{g}}}{2}\,v_{\mathrm{g}}^2\right)\right.$$
$$\left.\times\left(1-\mathrm{sinc}\left[2\sin\left(\frac{\theta}{2}\right)\frac{m_{\mathrm{g}}\,v_{\mathrm{g}}\,|\boldsymbol{x}|}{\hbar}\right]\right)\right\rangle_{v_{\mathrm{g}}}$$

scattering cross section

Decoherence by endogenous heat radiation

heating stage in front of interferometer



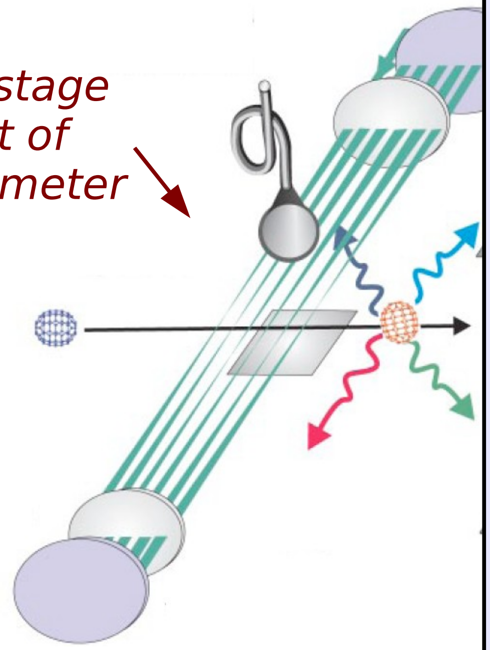
expected contrast reduction

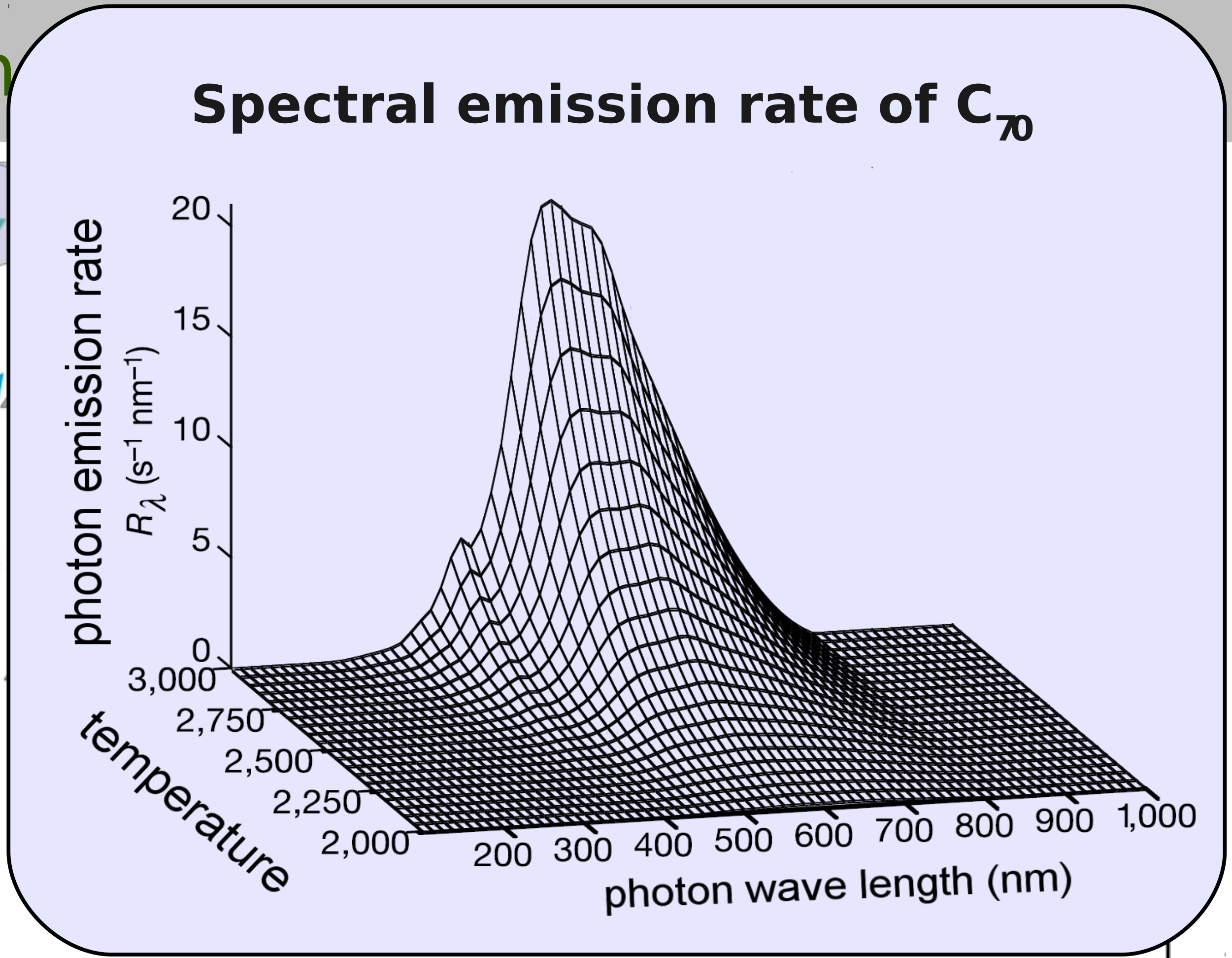
$$V = V_0 \ \exp\left[-\int_0^{2L/v} \mathrm{d}t \int_0^{\infty} \mathrm{d}\lambda \ R_\lambda\left(\lambda, T\left(t\right)\right)\left\{1 - \mathrm{sinc}\left(2\pi \frac{d}{\lambda}\frac{L - |vt - L|}{L_\mathrm{T}}\right)\right\}\right]$$

spectral emission rate of heat photons

"which-way" resolving power of a photon with wave length λ

Decoherence by en





expected contrast reduction

$$V = V_0 \, \exp\left[- \int_0^{2L/v} \mathrm{d}t \int_0^{\infty} \mathrm{d}\lambda \; R_\lambda\left(\lambda, T(t)\right) \left\{ 1 - \mathrm{sinc}\left(2\pi \frac{d}{\lambda} \frac{L - |vt - L|}{L_{\mathrm{T}}} \right) \right\} \right]$$

spectral emission rate of heat photons

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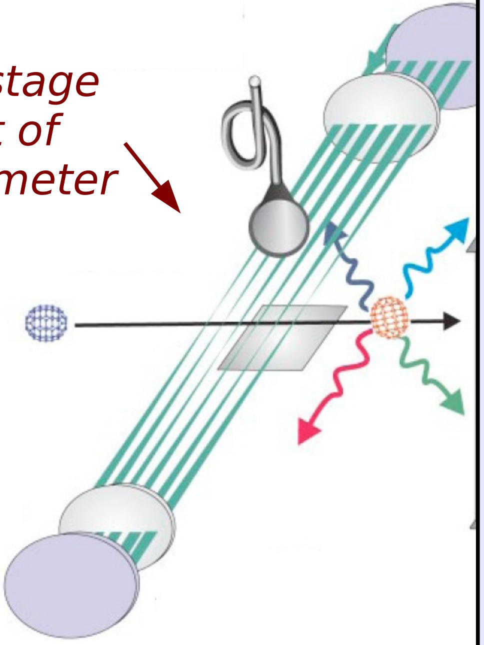
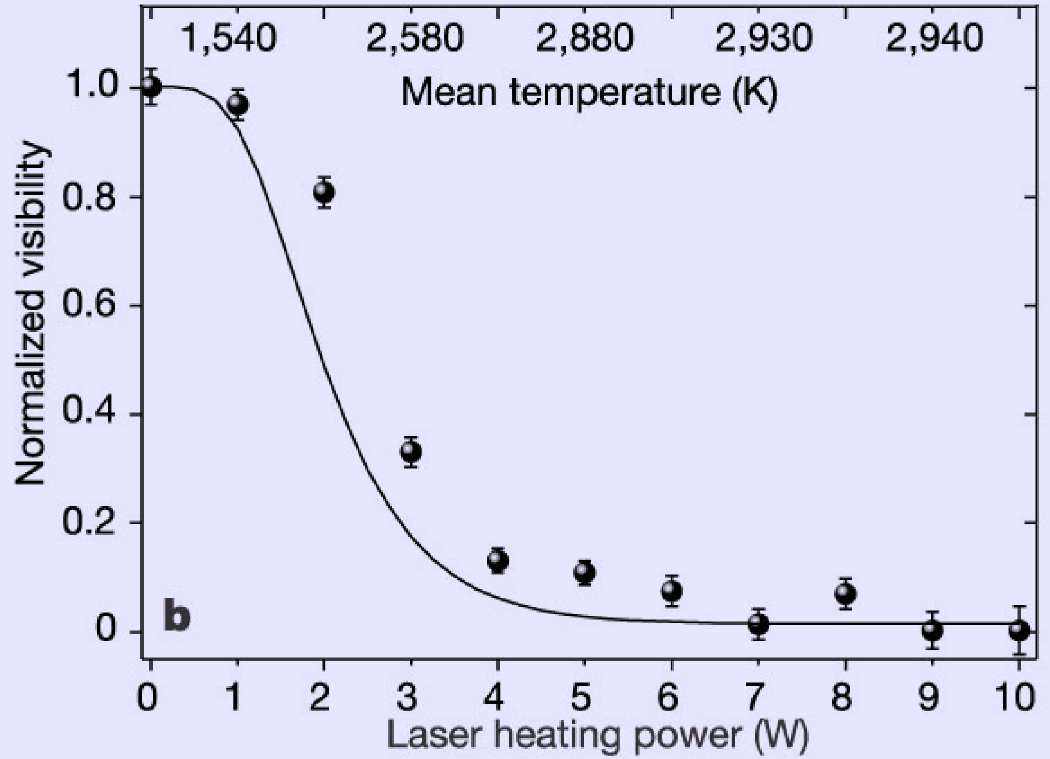
Decoherence by en



heating stage in front of interferometer

Experiment & theory



expected contrast reduction

$$V = V_0 \exp\left[-\int_0^{2L/v} \mathrm{d}t \int_0^{\infty} \mathrm{d}\lambda \; R_\lambda\left(\lambda, T(t)\right) \left\{ 1 - \mathrm{sinc}\left(2\pi \frac{d}{\lambda} \frac{L - |vt - L|}{L_\mathrm{T}} \right) \right\} \right]$$

spectral emission rate of heat photons

“which-way” resolving power of a photon with wave length λ

Decoherence by collisions with a background gas

expected contrast reduction

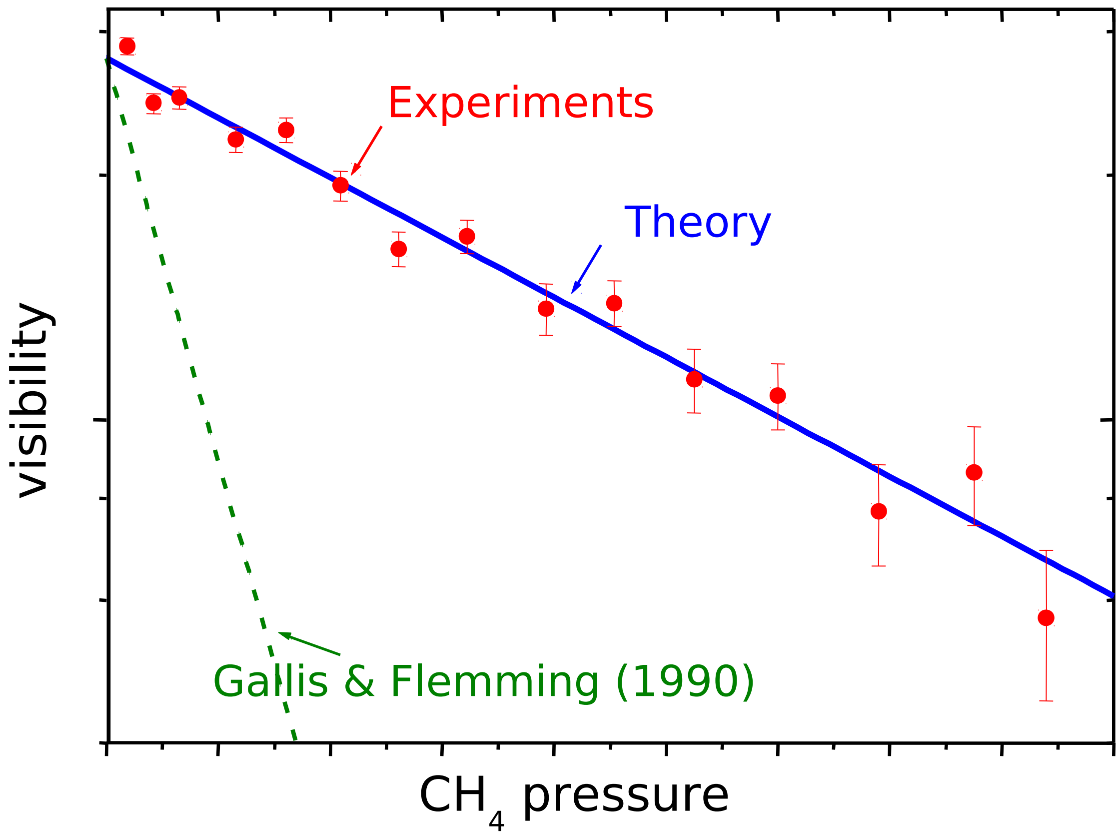
$$V = V_0 \exp\left(-\frac{2L\sigma_{\text{eff}}}{k_B T} p\right)$$

with

$$\sigma_{\text{eff}}(v_m) = \frac{4\pi\Gamma(9/10)}{5\sin(\pi/5)} \left(\frac{3\pi C_6}{2\hbar}\right)^{2/5} \frac{\tilde{v}_g^{3/5}}{v_m} \times \left\{1 + \frac{1}{5}\left(\frac{\tilde{v}_g}{v_m}\right)^2 + \mathcal{O}\left(\frac{\tilde{v}_g}{v_m}\right)^4\right\}$$

in case of van der Waals-WW

$$U_{\text{int}} = -C_6/r^6$$

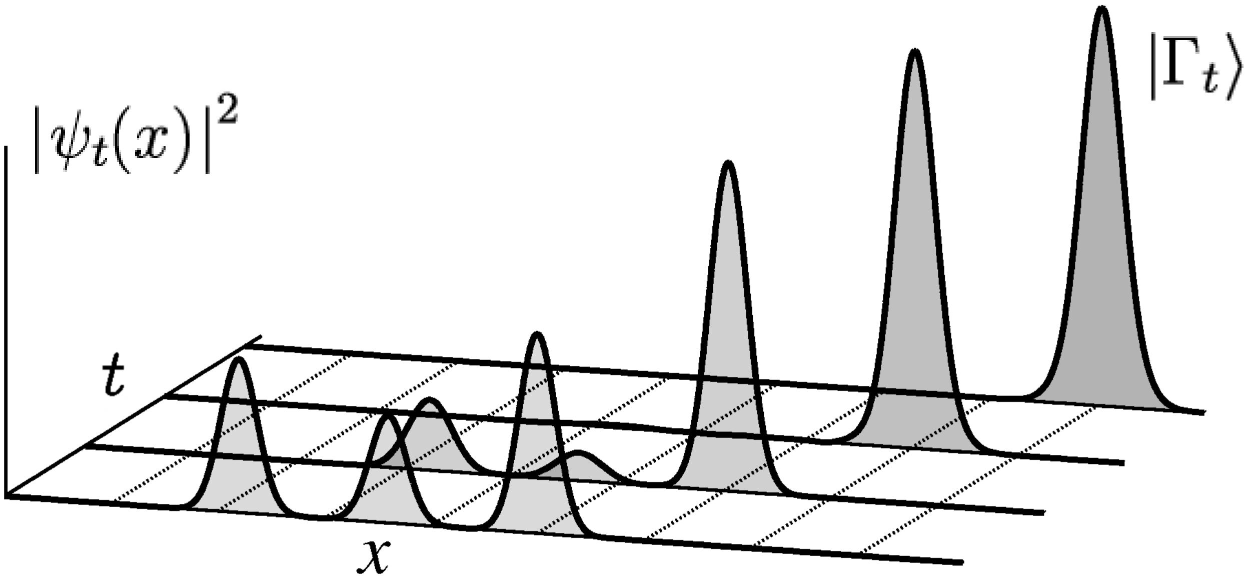


Pointer states of collisional decoherence

$$\rho_0 \xrightarrow{t \gg t_{\mathrm{dec}}} \rho_t \simeq \int \mathrm{d}\Gamma_0 \, \mathrm{Prob}(\Gamma_0|\rho_0) \, |\Gamma_t\rangle\langle\Gamma_t|$$

Born's rule *pointer basis*

$|\Gamma_t\rangle$: exponentially localized solitons
evolve on classical trajectories



[Rigo, Gisin (2000)]

Overview

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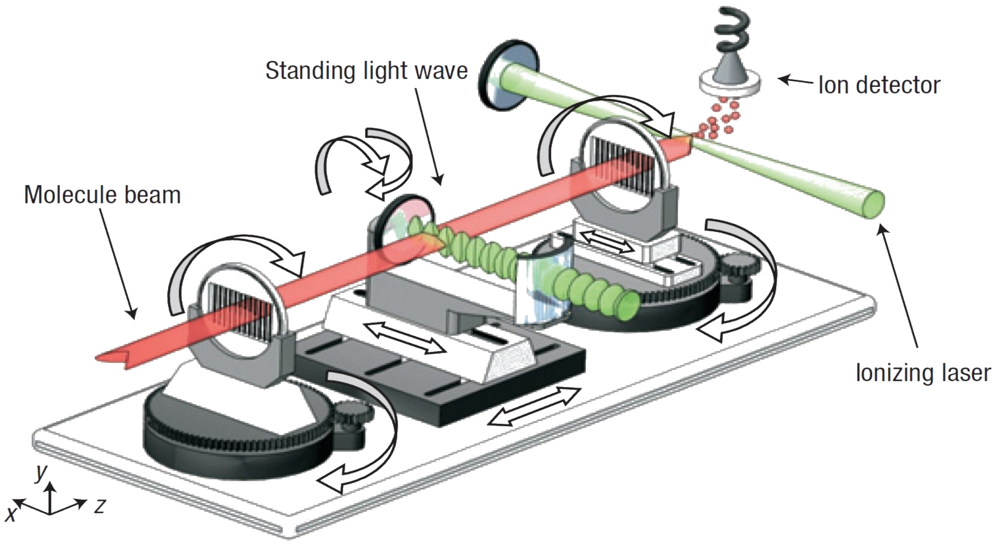
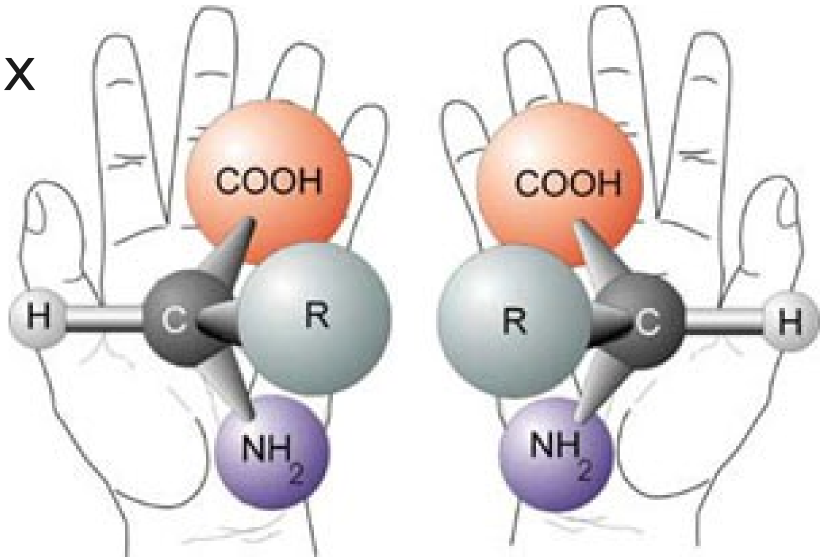
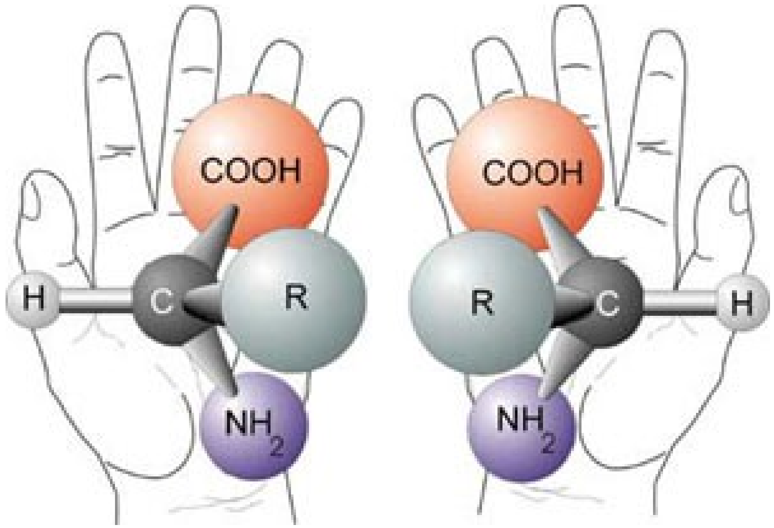


2. Hund's paradox of molecular chirality



Hund's paradox of molecular chirality

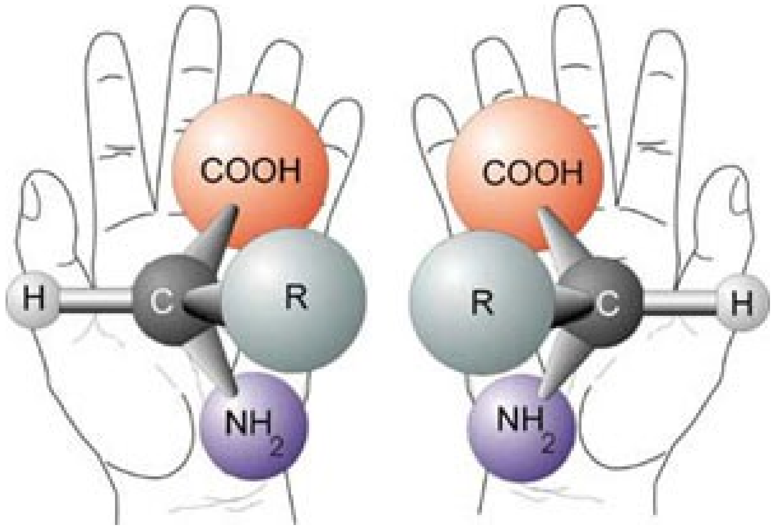


Friedrich Hund (1927)

Why are many molecules found in a chiral configuration?

—in spite of the parity invariance of their hamiltonian?

Hund's paradox of molecular chirality

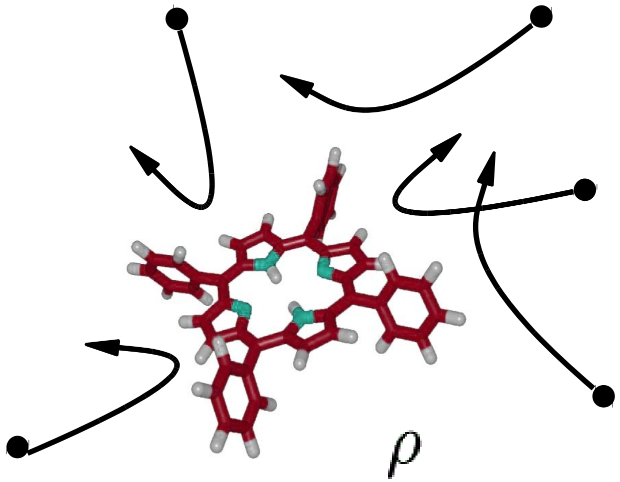


Friedrich Hund (1927)

Why are many molecules found in a chiral configuration?

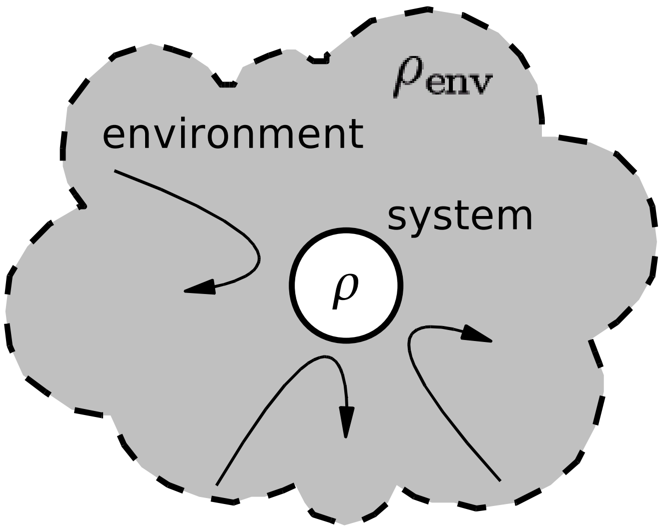
—in spite of the parity invariance of their hamiltonian?

Effect of an *achiral* gas environment on the configuration & orientation state?



non-perturbative quantum master equation required

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho \overset{?}{=} \mathcal{L}\rho$$



Idea:

Don't start with the Schrödinger equation for the total system

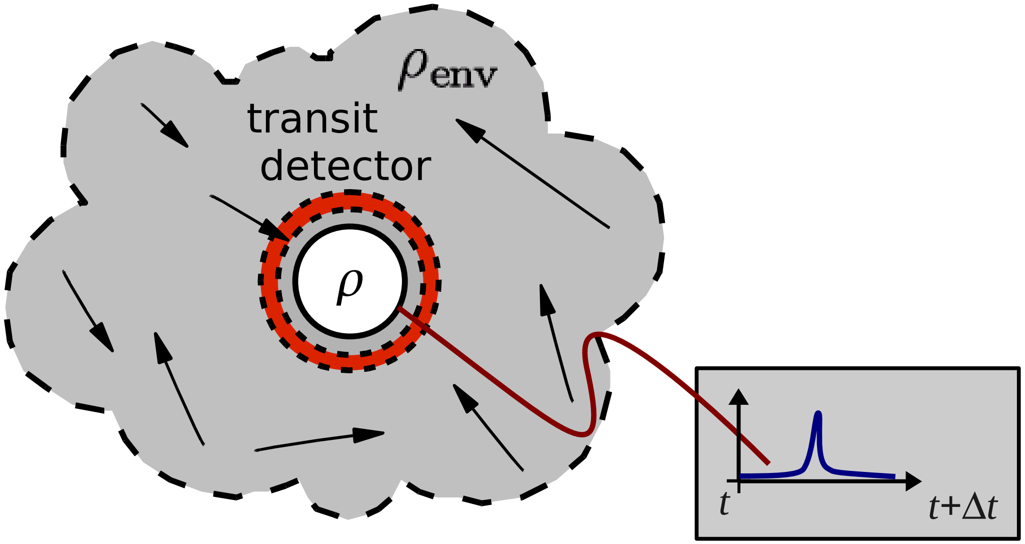
but put the Markov assumption (“memoryless environment”)

as the central premise!

Monitoring approach

rate operator Γ and
scattering operator S
characterize events

$(S = I + iT)$



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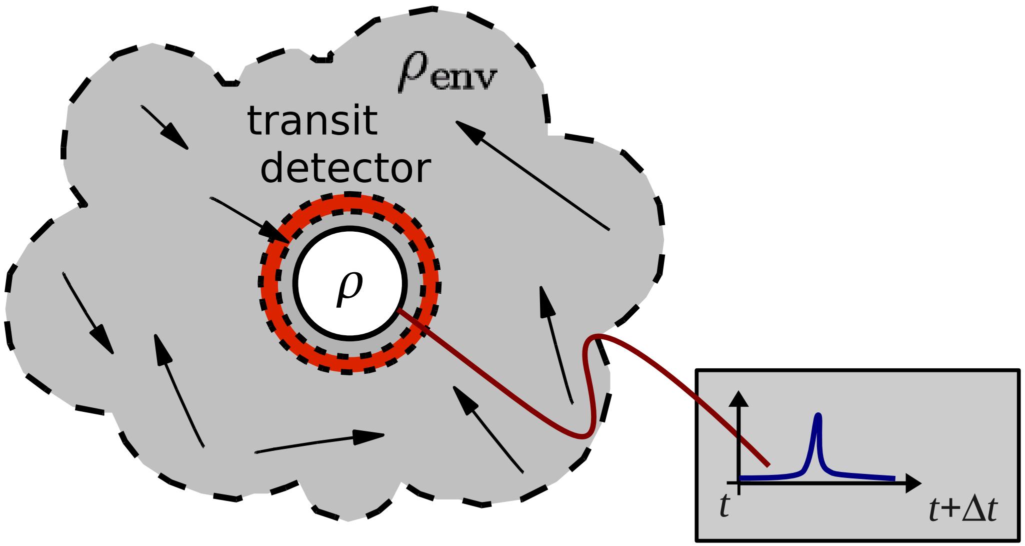

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\rho \;=\; & \frac{1}{i\hbar}[H,\rho] + i\,\mathrm{Tr}_{\mathrm{env}}\Big(\big[\Gamma^{1/2}\mathrm{Re}(T)\Gamma^{1/2}, \rho\otimes\rho_{\mathrm{env}}\big]\Big) \\
& + \mathrm{Tr}_{\mathrm{env}}\Big(T\Gamma^{1/2}[\rho\otimes\rho_{\mathrm{env}}]\Gamma^{1/2}T^\dagger\Big) \\
& - \frac{1}{2}\,\mathrm{Tr}_{\mathrm{env}}\Big(\Gamma^{1/2}T^\dagger T\,\Gamma^{1/2}[\rho\otimes\rho_{\mathrm{env}}]\Big) \\
& - \frac{1}{2}\,\mathrm{Tr}_{\mathrm{env}}\Big([\rho\otimes\rho_{\mathrm{env}}]\,\Gamma^{1/2}T^\dagger T\,\Gamma^{1/2}\Big)
\end{aligned}
$$

Monitoring approach

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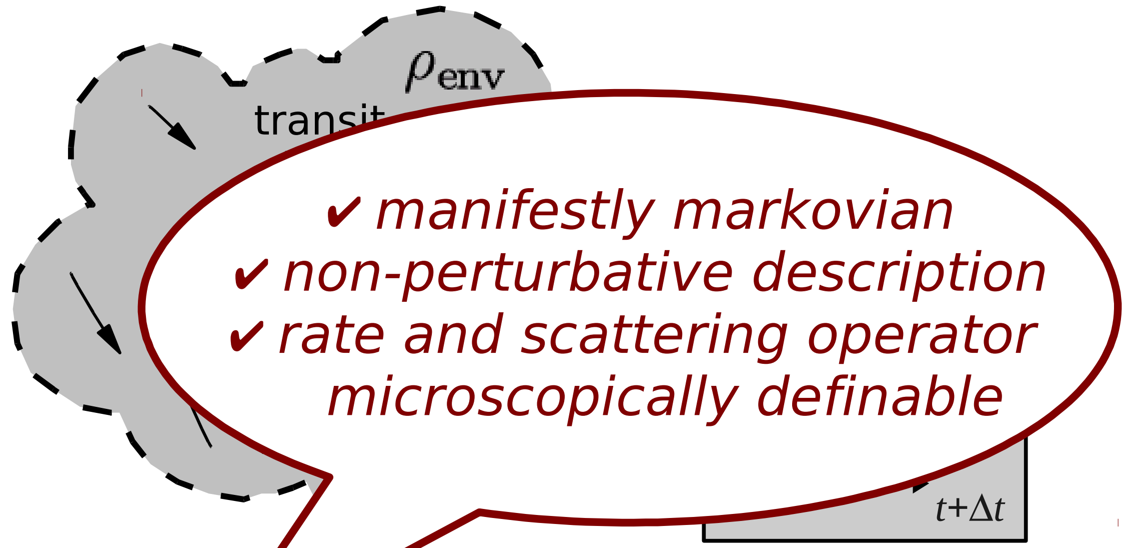
$(S = I + iT)$



✔ *manifestly markovian*
✔ *non-perturbative description*
✔ *rate and scattering operator microscopically definable*

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\rho \;=\; & \frac{1}{i\hbar}[H,\rho] + i\,\mathrm{Tr}_{\mathrm{env}}\Big(\big[\Gamma^{1/2}\mathrm{Re}(T)\Gamma^{1/2}, \rho\otimes\rho_{\mathrm{env}}\big]\Big) \\
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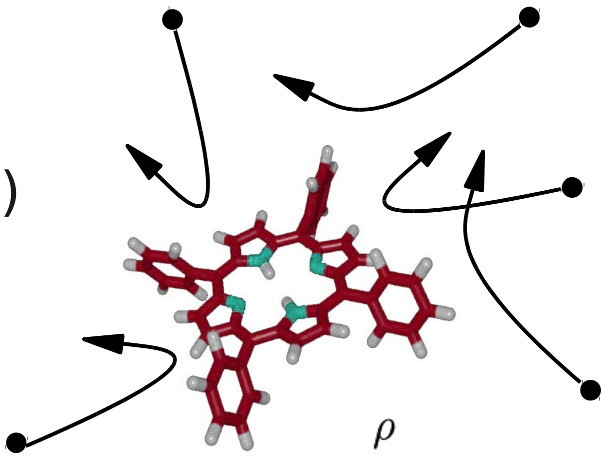
& - \frac{1}{2}\,\mathrm{Tr}_{\mathrm{env}}\Big([\rho\otimes\rho_{\mathrm{env}}]\,\Gamma^{1/2}T^{\dagger}T\,\Gamma^{1/2}\Big)
\end{aligned}
$$

microscopically realistic choice

$\Gamma =$ (gas current density) x (cross section)

$S =$ (multi-channel S-Matrix)



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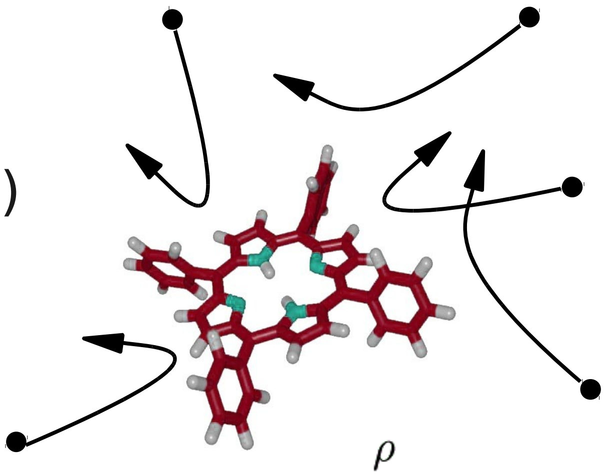


yields:

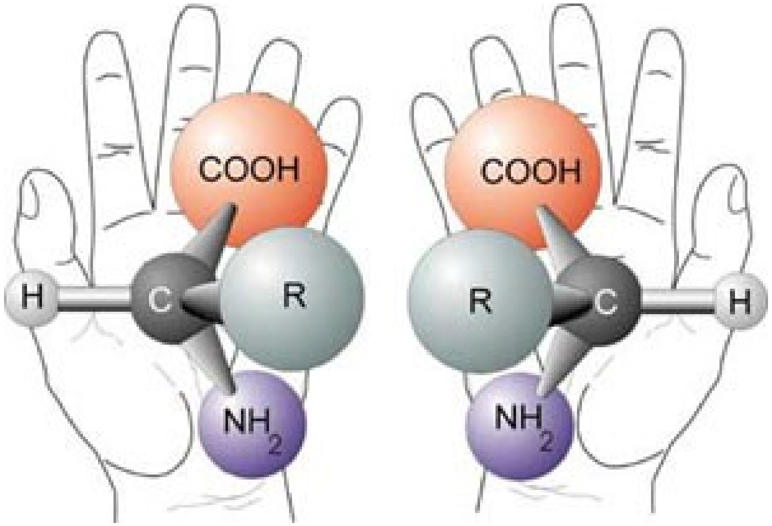
$$M_{\alpha\beta}^{\alpha_0\beta_0} = n_{\rm gas} \int \mathrm{d}\Omega \left\langle v_{\rm out}\, f_{\alpha\alpha_0}\Big(\cos\theta; \frac{m}{2} v_{\rm in}^2\Big)\, f^*_{\beta\beta_0}\Big(\cos\theta; \frac{m}{2} v_{\rm in}^2\Big)\right\rangle_{v_{\rm in}}$$

$$\partial_t \rho_{\alpha\beta} = -i\omega_{\alpha\beta}\,\rho_{\alpha\beta} + \sum_{\substack{\alpha_0\beta_0 \\ \omega_{\alpha\alpha_0}=\omega_{\beta\beta_0}}} \rho_{\alpha_0\beta_0}\, M_{\alpha\beta}^{\alpha_0\beta_0} - \frac{1}{2} \sum_{\substack{\alpha_0 \\ \omega_\alpha=\omega_{\alpha_0}}} \rho_{\alpha_0\beta} \sum_{\gamma} M_{\gamma\gamma}^{\alpha_0\alpha} - \frac{1}{2} \sum_{\substack{\beta_0 \\ \omega_\beta=\omega_{\beta_0}}} \rho_{\alpha\beta_0} \sum_{\gamma} M_{\gamma\gamma}^{\beta\beta_0}$$

(extends Dümcke 1985)

Hund's paradox of molecular chirality

Effect of an *achiral* gas environment



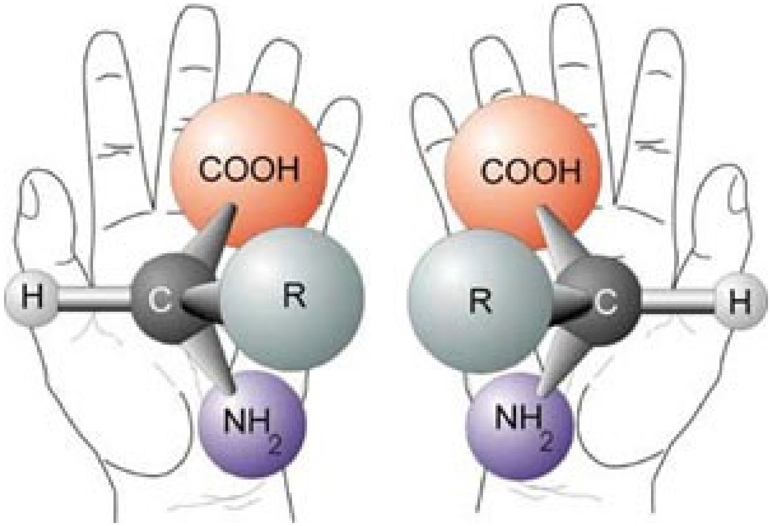
- $|L\rangle + \mathrm{e}^{i\varphi}|R\rangle$ decay with decoherence rate

$$\gamma \;=\; n_{\mathrm{gas}} \left\langle v \underbrace{\int \frac{\mathrm{d}\boldsymbol{n}\,\mathrm{d}\boldsymbol{n}_0}{8\pi} \left| f^{(L)}_{\alpha,\alpha_0}(v\,\boldsymbol{n}, v\,\boldsymbol{n}_0) - f^{(R)}_{\alpha,\alpha_0}(v\,\boldsymbol{n}, v\,\boldsymbol{n}_0) \right|^2}_{\text{“decoherence cross section”}} \right\rangle_{v,\alpha,\alpha_0}$$

“decoherence cross section”

Hund's paradox of molecular chirality

Effect of an *achiral* gas environment



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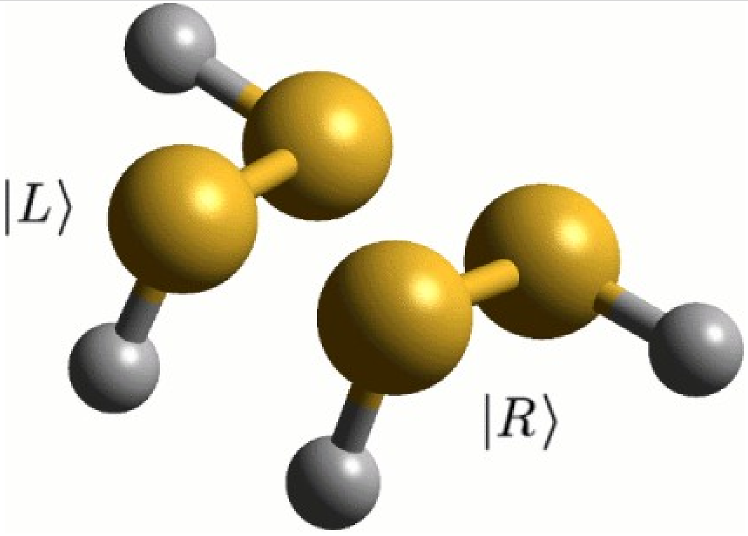
- *only* the chiral states $|L\rangle$ and $|R\rangle$ exhibit a quantum-Zeno-like stabilization $\sim\omega^2/\gamma$ against tunneling and decay if $\gamma \gg \omega$

Harris, Stodolsky (1978)

Hund's paradox of molecular chirality

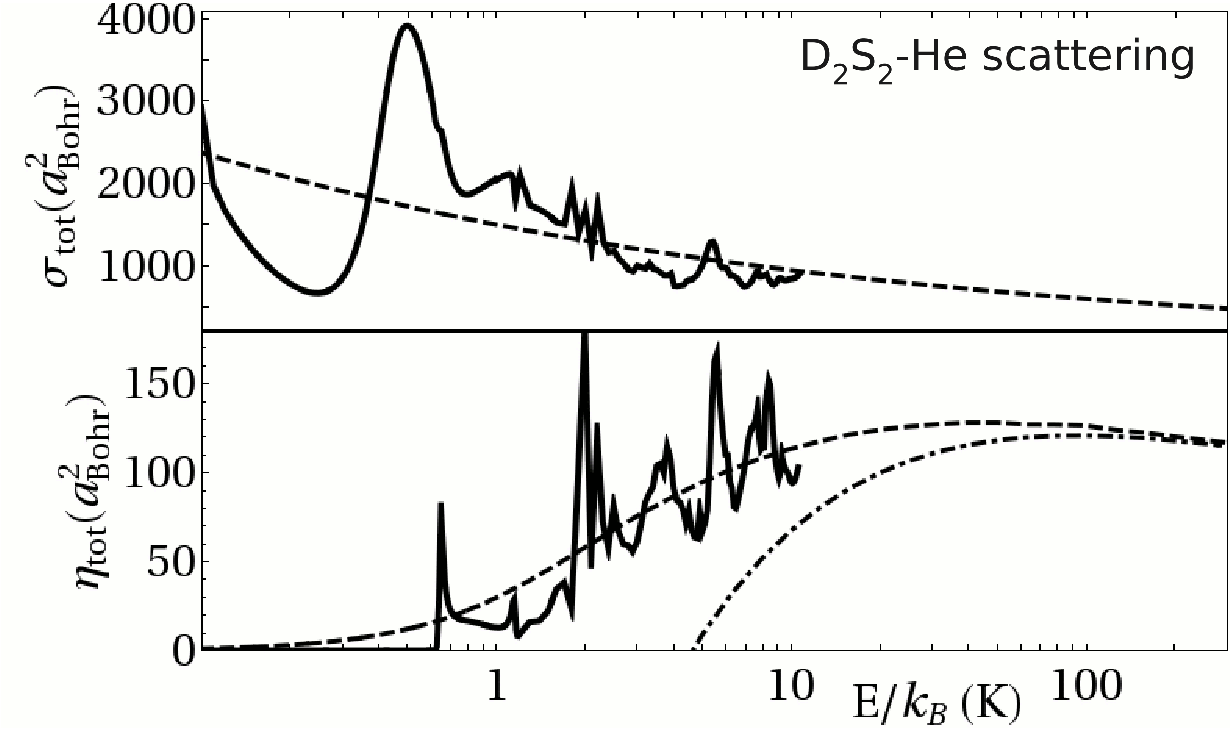


D_2S_2 tunnels with 176 Hz in vacuum

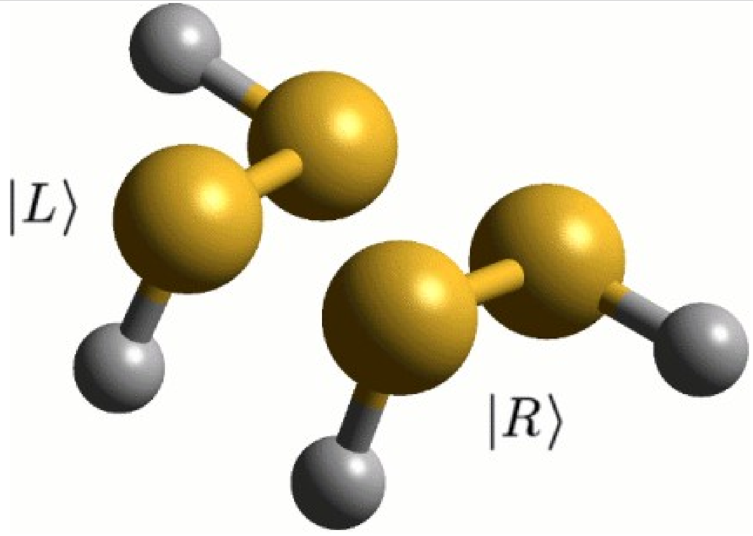
The stabilization effect is dominated by a higher order contribution to the van der Waals interaction described by the EQED tensor $A_{j,k\ell}(i\omega)$

scattering cross section

decoherence cross section



Hund's paradox of molecular chirality



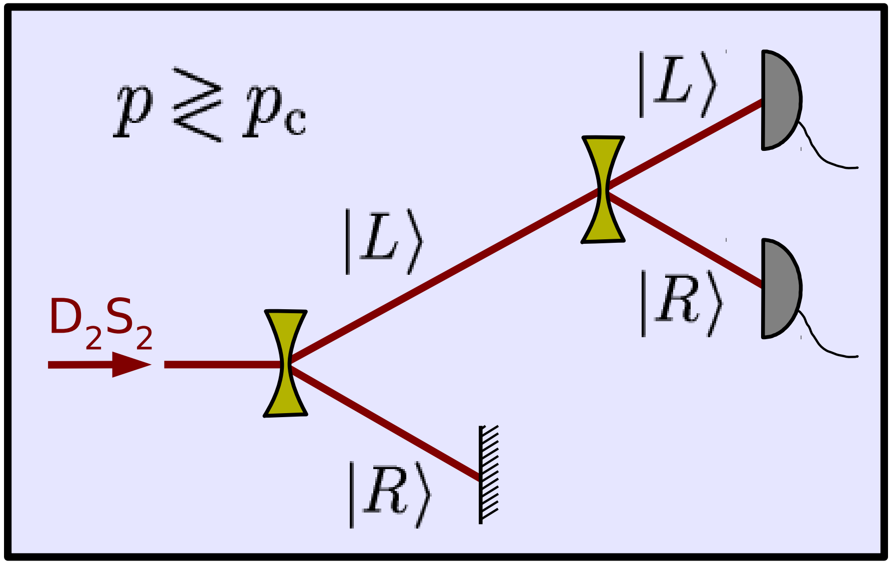
D_2S_2 tunnels with 176 Hz in vacuum

The stabilization effect is dominated by a higher order contribution to the van der Waals interaction described by the EQED tensor $A_{j,k\ell}(i\omega)$

critical pressure in 300K He atmosphere:

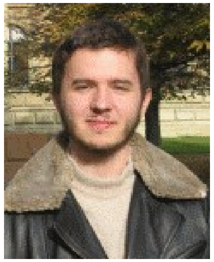
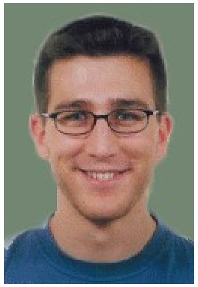
$$p_c = 1.6 \times 10^{-5} \text{mbar}$$

... allows one to observe the chiral stabilization in an optical Stern-Gerlach type setup



acknowledgments

theory

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LMU Munich
U Vienna

experiments



Markus Arndt
Lucia Hackermüller,
Stefan Gerlich et al *U Vienna*

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