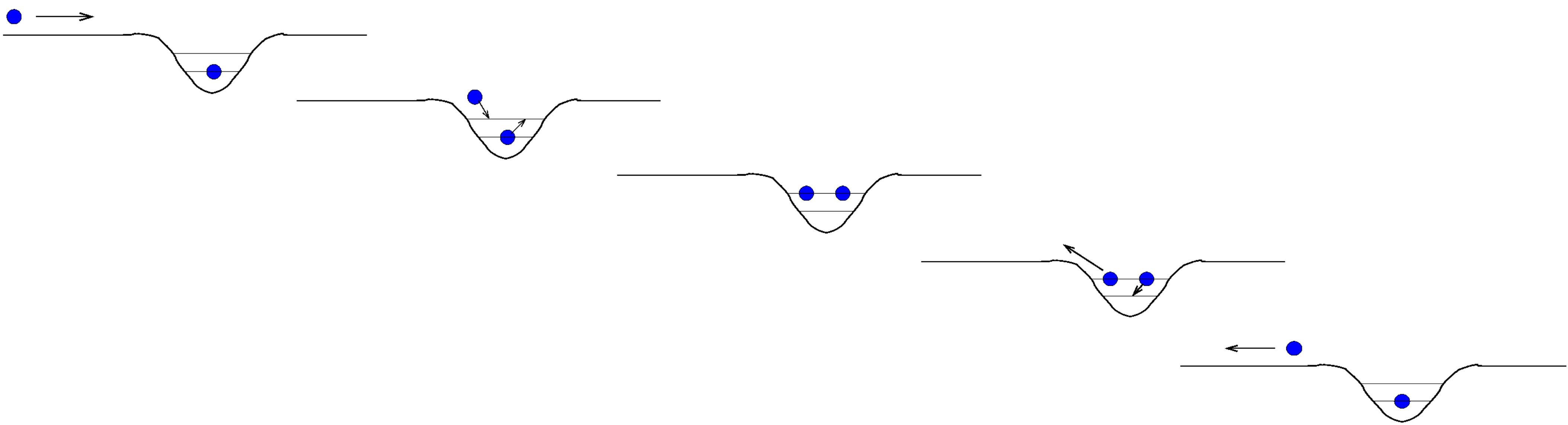
Dielectronic recombination in the time domain



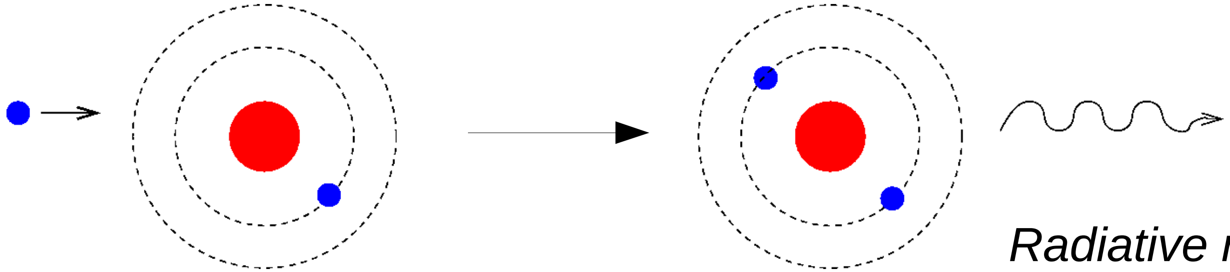
Control of Ultrafast Quantum Phenomena

Nordita, Stockholm

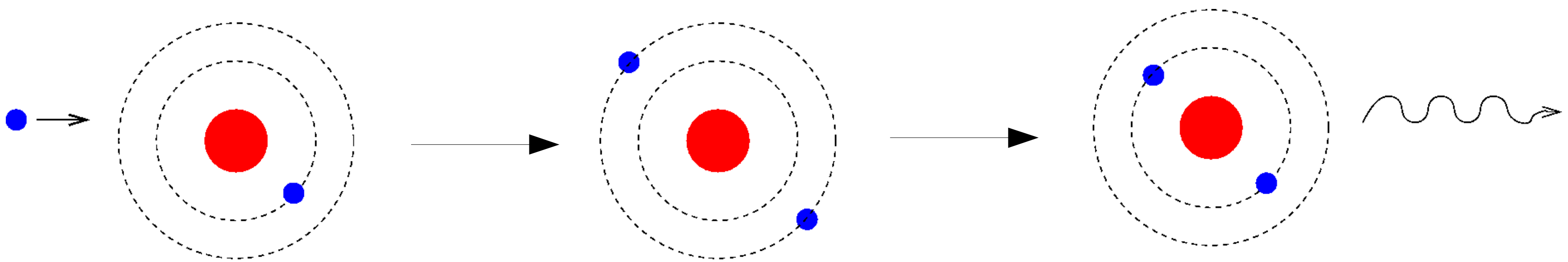
OSLO AND AKERSHUS UNIVERSITY COLLEGE OF APPLIED SCIENCES

Sølve Selstø, Oslo and Akershus University College of Applied Sciences

Recombination



Radiative recombination



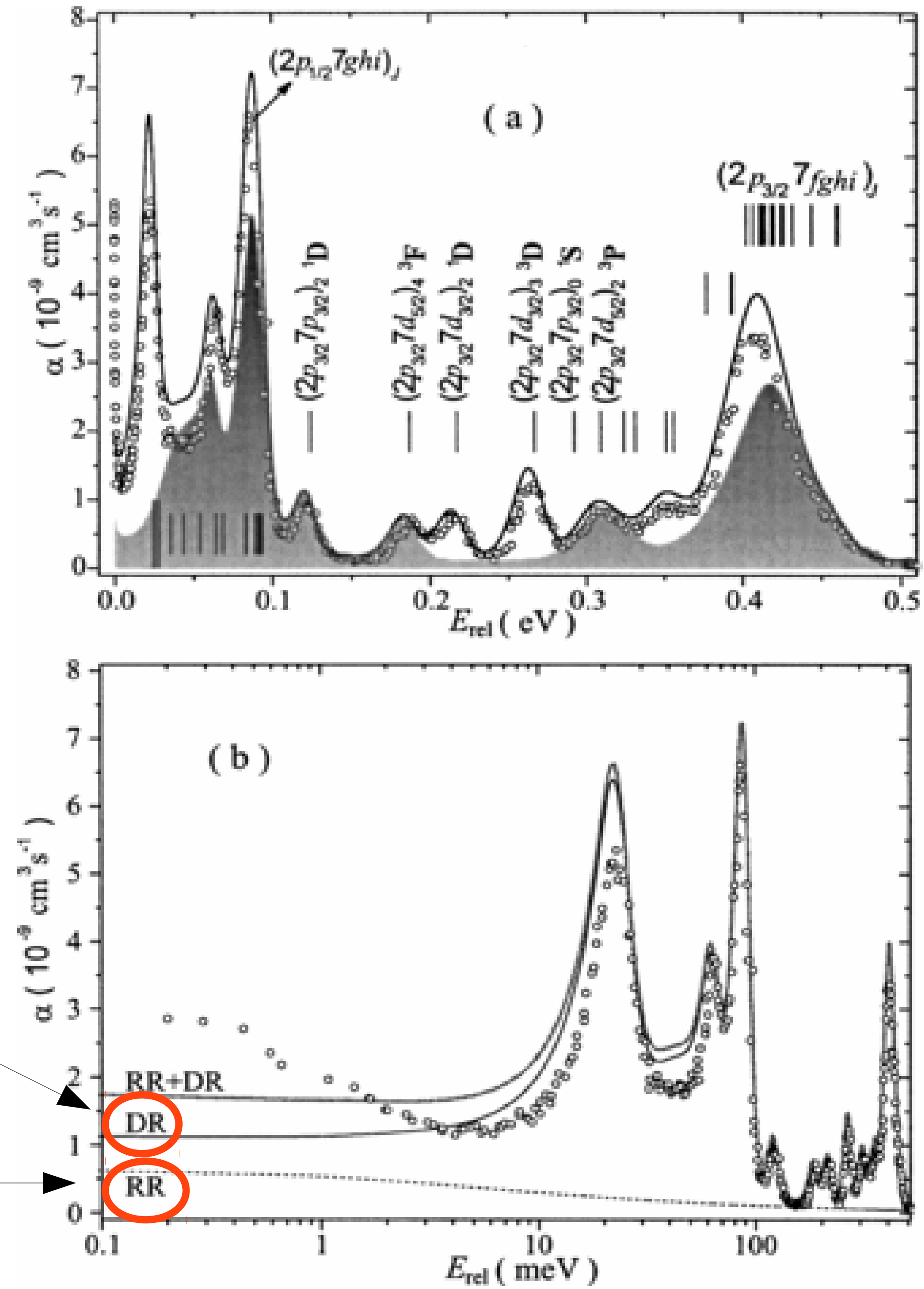
Dielectronic recombination

Dielectronic recombination:

- *Time reverse of auto ionization (Auger decay)*
- *May increase the capture probability by orders of magnitude*
- *Dictated by the structure of the system*

Recombination

Na^{8+}

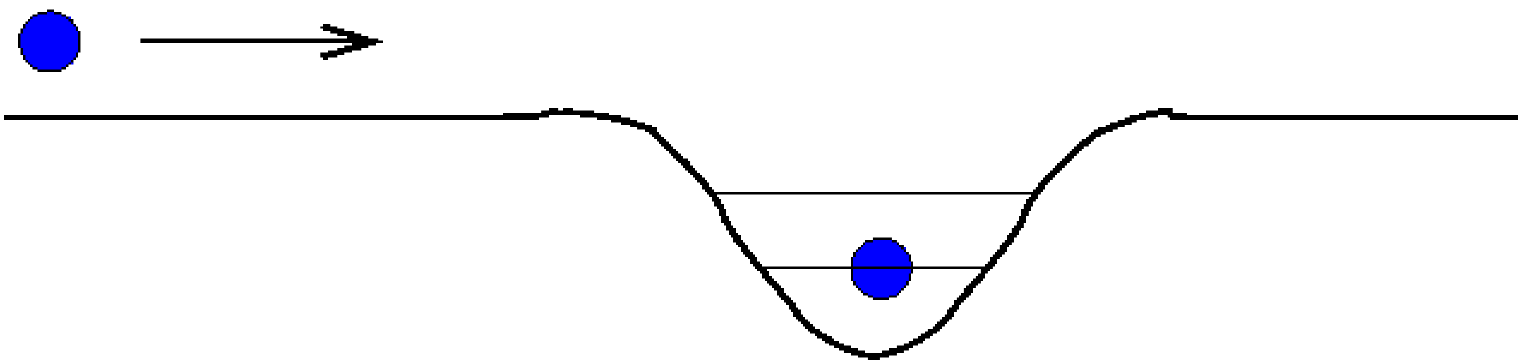


Dielectronic recombination

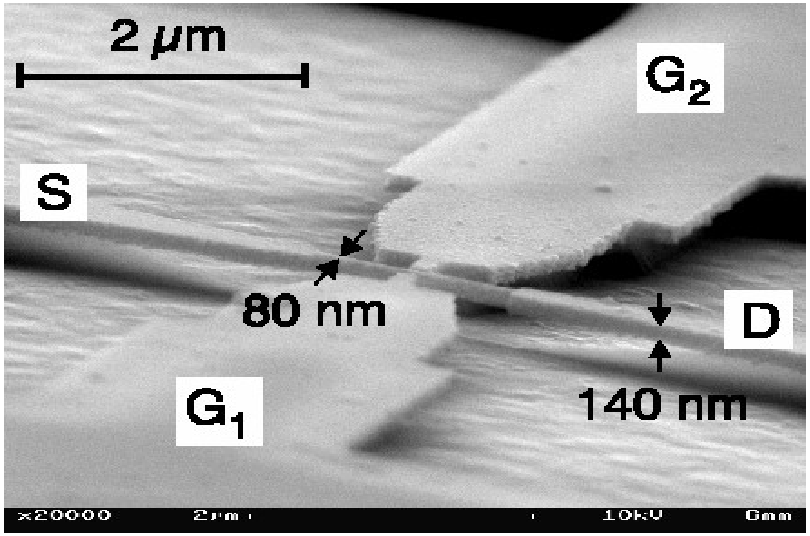
Radiative recombination

D Nicolić et al, *Phys. Rev. A* **70**, 062723 (2004)

The system

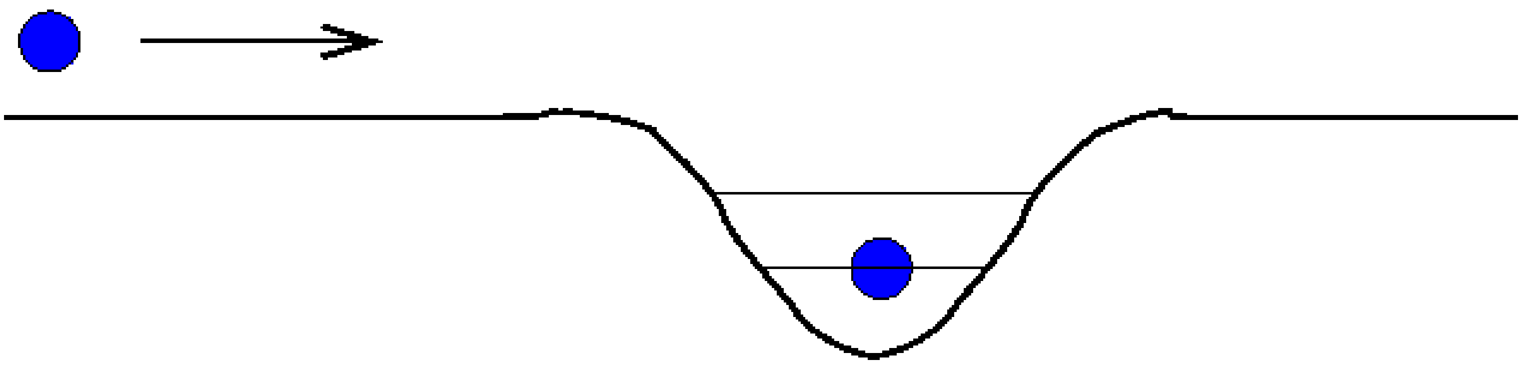


Possible realization: Quantum dot embedded in a quantum wire



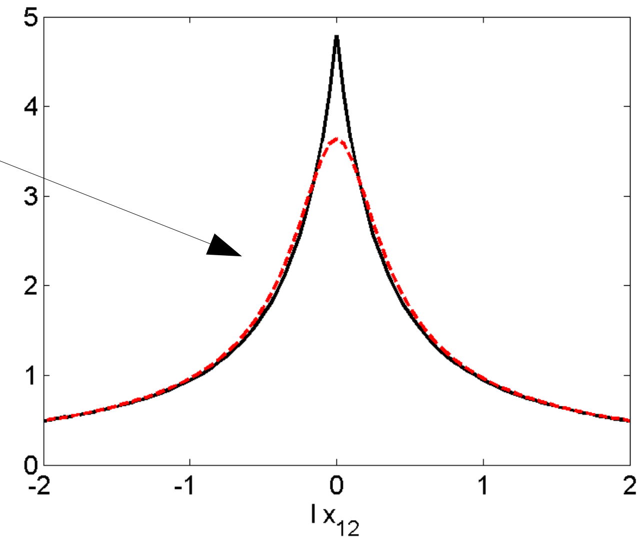
From the website of Dr. Bert Lorenz',
Ludwig-Maximilians-Universität München

The system



Interaction: Regularized **Coulomb interaction** with transversial degrees of freedom integrated out;

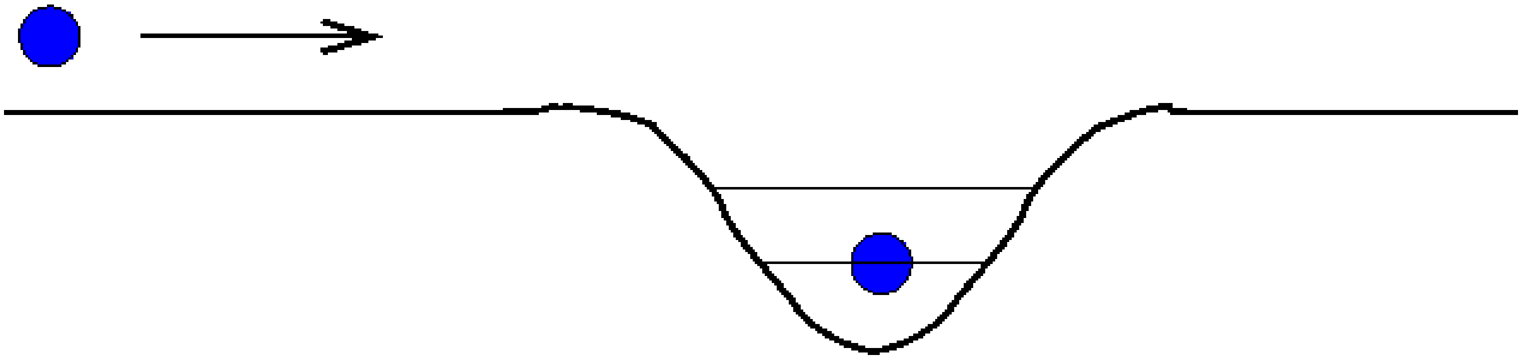
$$\overline{W}(x_{12}) = \frac{1}{\sqrt{x_{12}^2 + (l\delta)^2}}$$



Confining potential (quantum dot): **Gaussian**

$$\widetilde{V}(x) = -D_V \exp\left(-\frac{x^2}{\sigma_V^2}\right)$$

The system



Units

$$m^* = e = \hbar = \frac{1}{4\pi\varepsilon_r\varepsilon_0} = 1$$

For GaAs:

Length unit: ~ 10 nm

Energy unit: ~ 11 meV

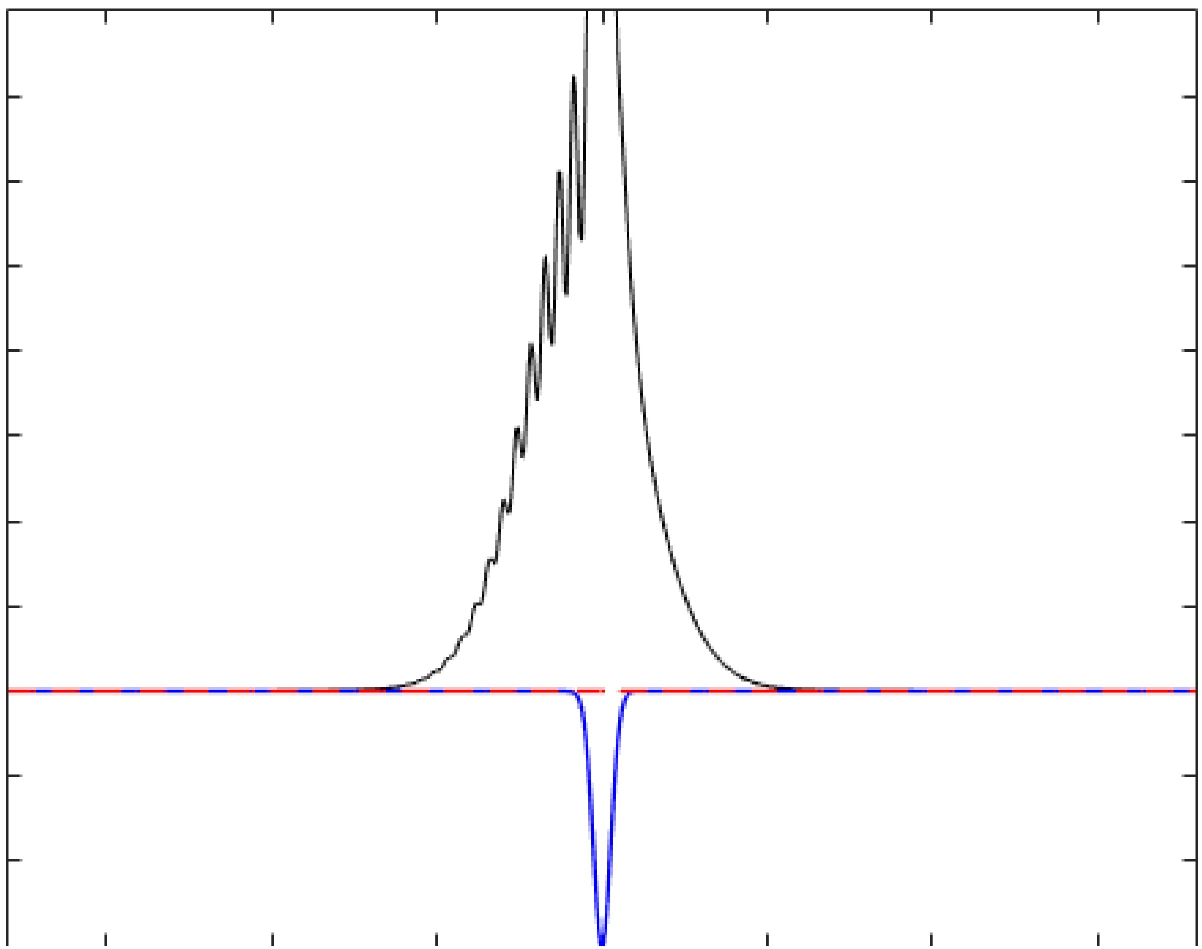
Time unit: ~ 60 fs

Resolve the dynamics by solving the relevant *dynamical* equations

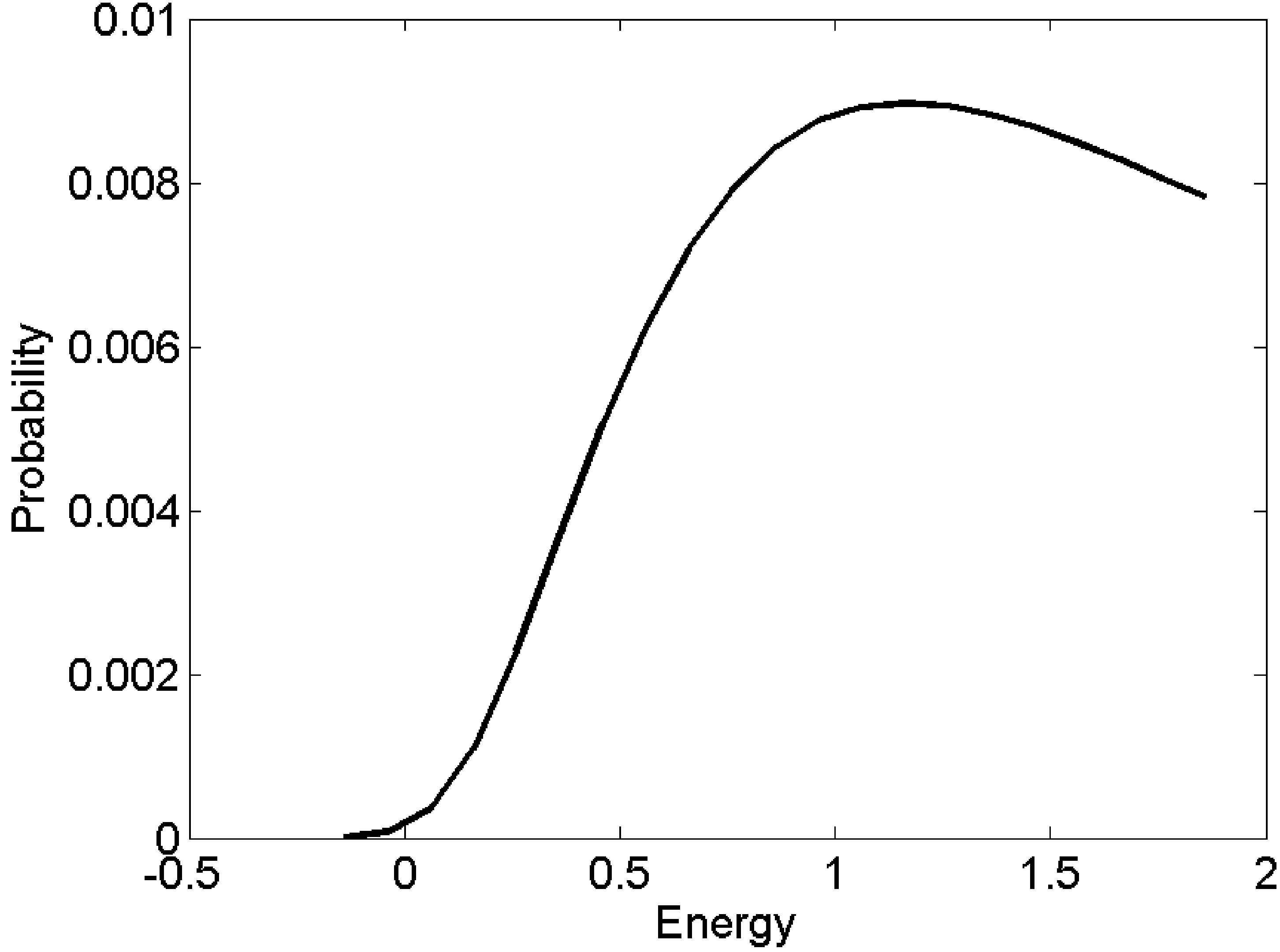
Resolve the dynamics by solving the relevant *dynamical* equations

Why explicit time dependence?

- This *is* a dynamical process; this is in fact the proper framework
- Examine the buildup and decay of the population of resonant states
- No need for scattering states
- It's fun

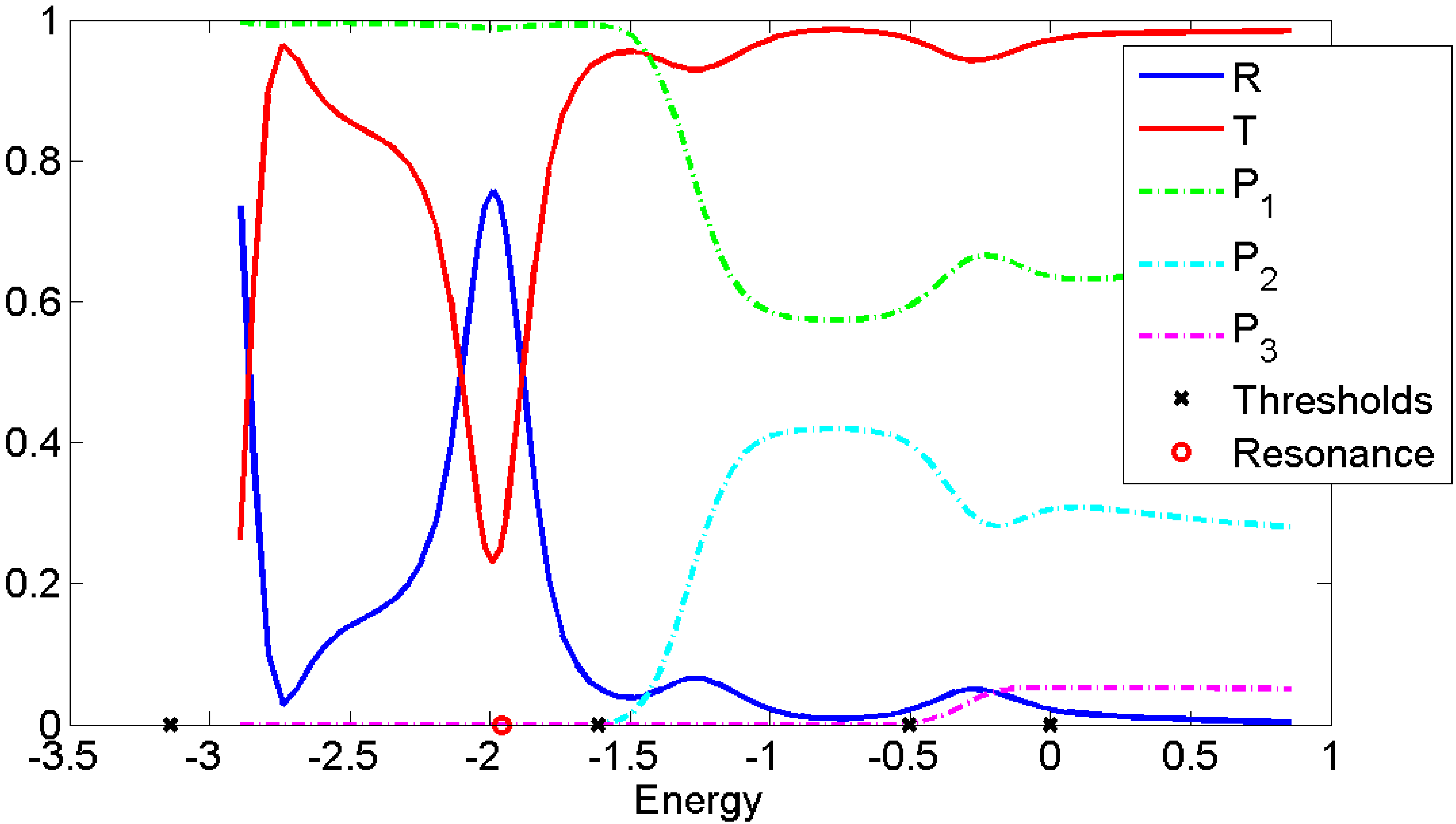


Finite probability for «knockout»/«Ionization»



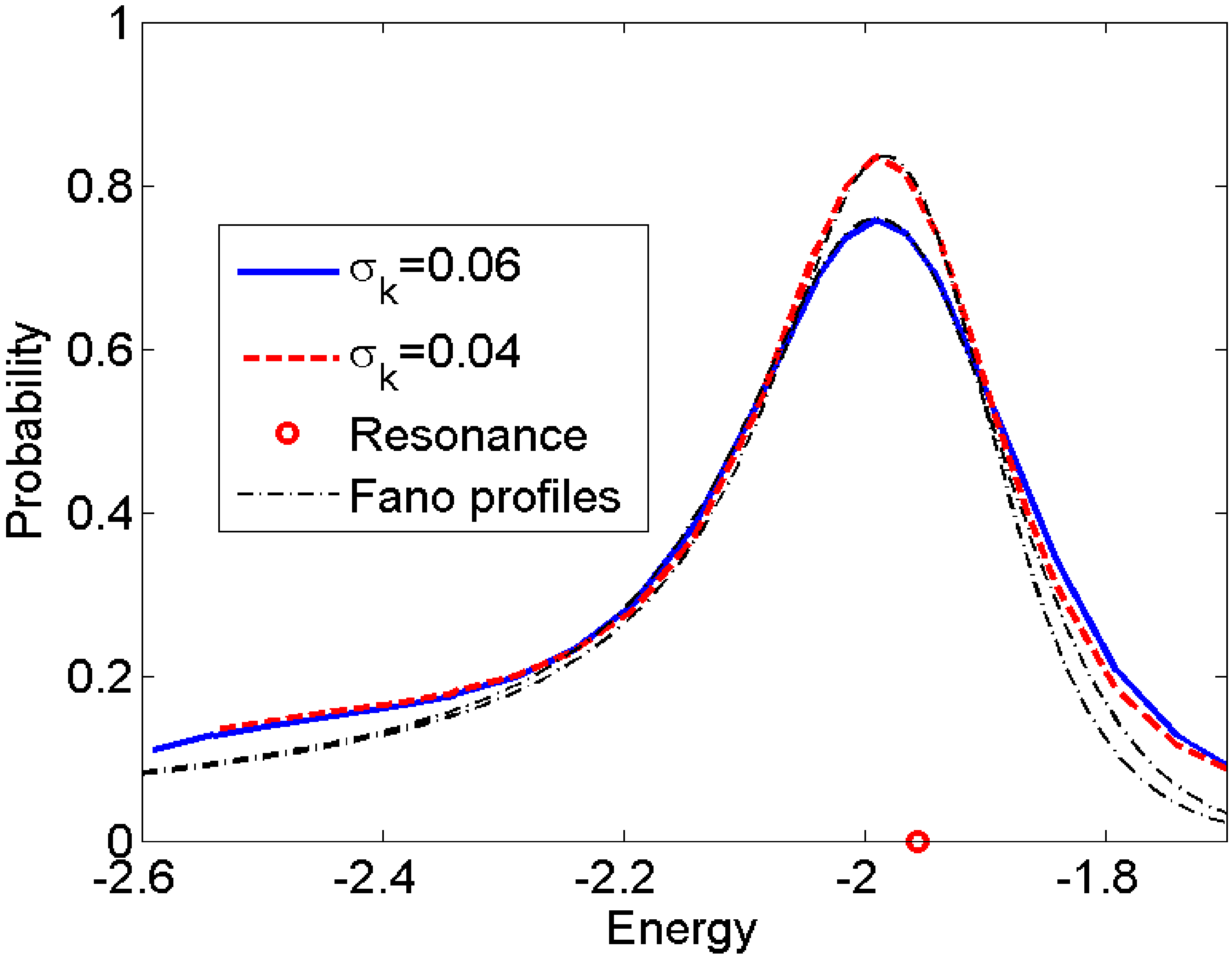
S. S., *J. Phys. Cond. Matt.* **25** 315802 (2013)

Final probabilities



S. S., *J. Phys. Cond. Matt.* **25** 315802 (2013)

The resonance peak in the reflection coefficient

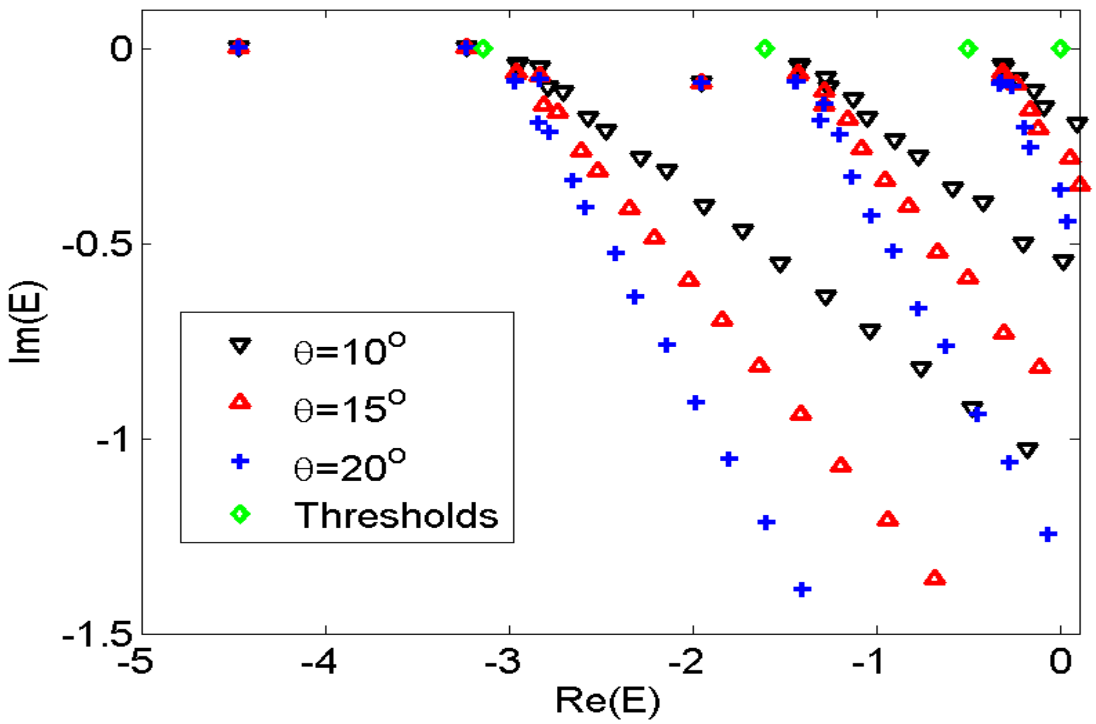


q=-4.5

S. S., *J. Phys. Cond. Matt.* **25** 315802 (2013)

Identify doubly excited states by complex rotation,

$x \rightarrow x\, e^{i\theta},\ \ 0 < \theta < 45°$



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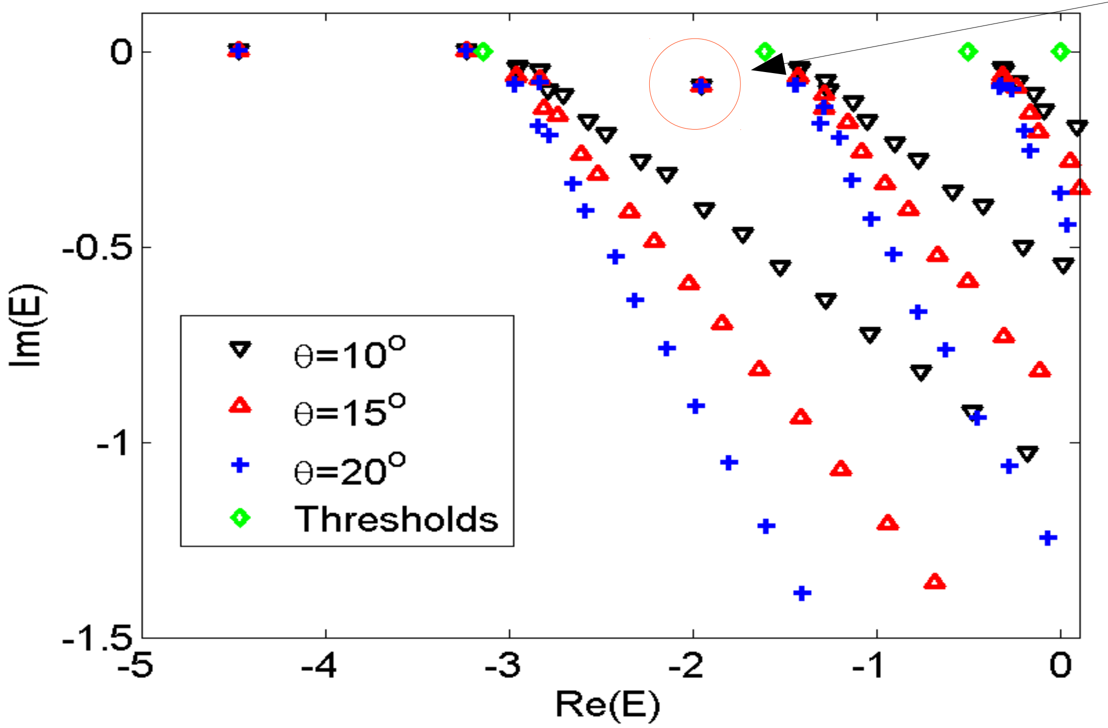


$E = E_{res} - i\,\Gamma_{res}/2$

Position

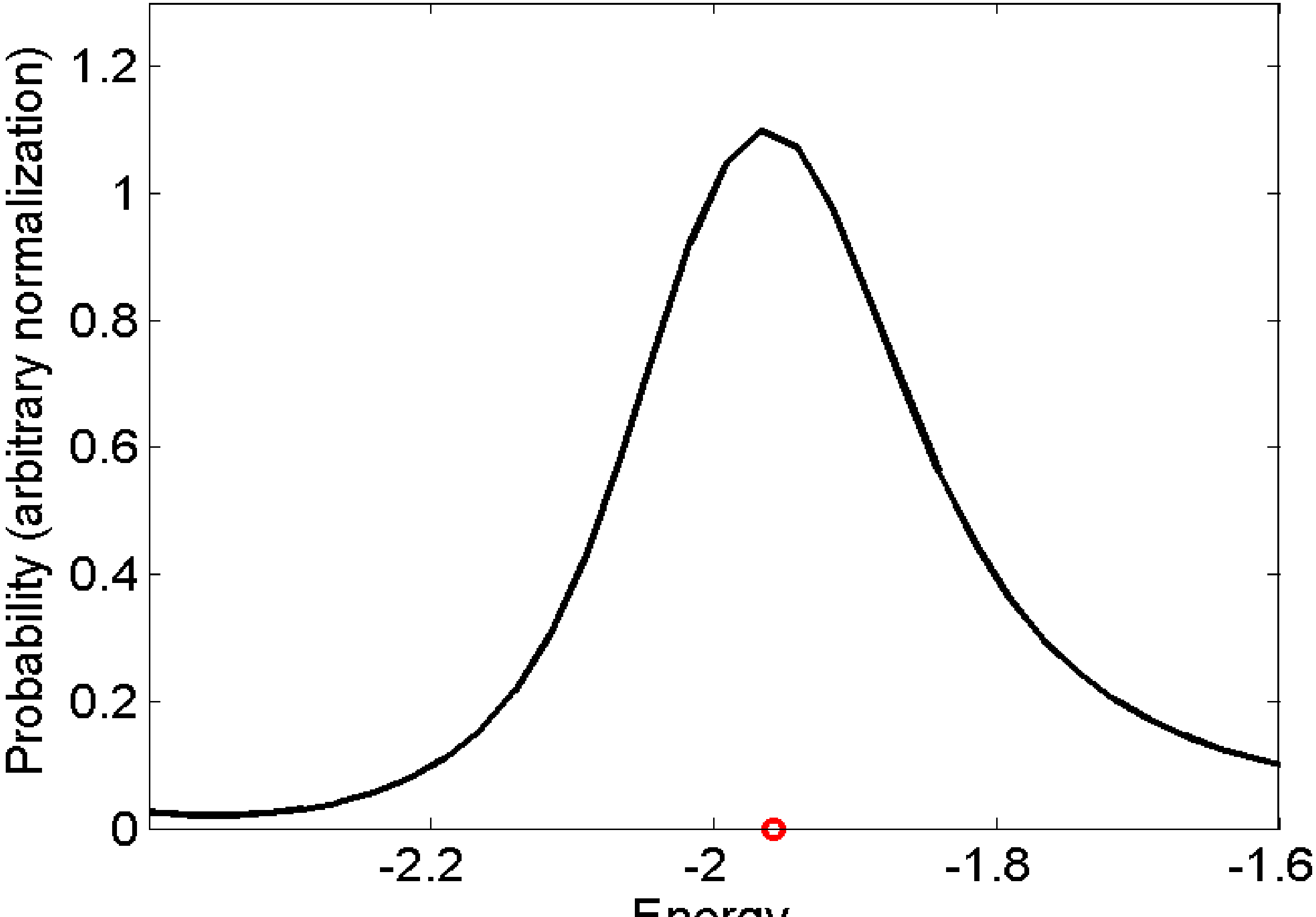
Width

$E_{res} = -1.96$

$\Gamma_{res} = \; 0.179$

Photon Capture

Guess what: The resonance dominates

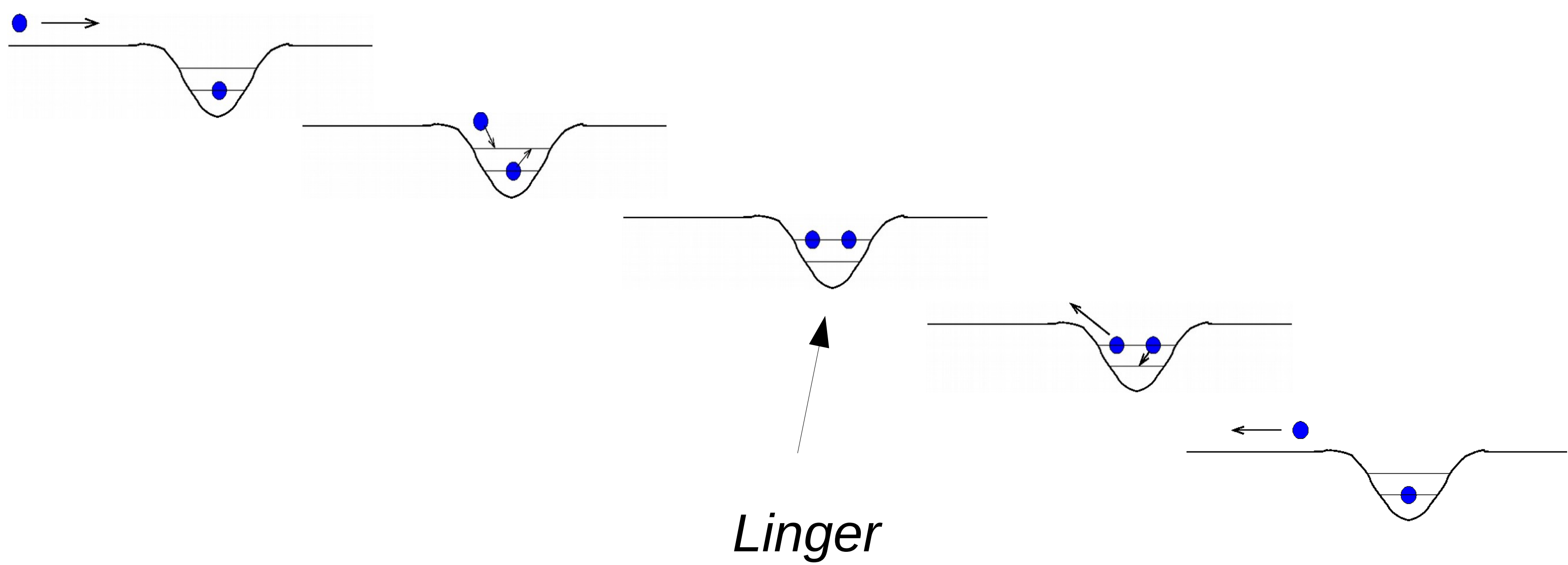


S. S., *J. Phys. Cond. Matt.* **25** 315802 (2013)

Photon Capture

Guess what: The resonance dominates

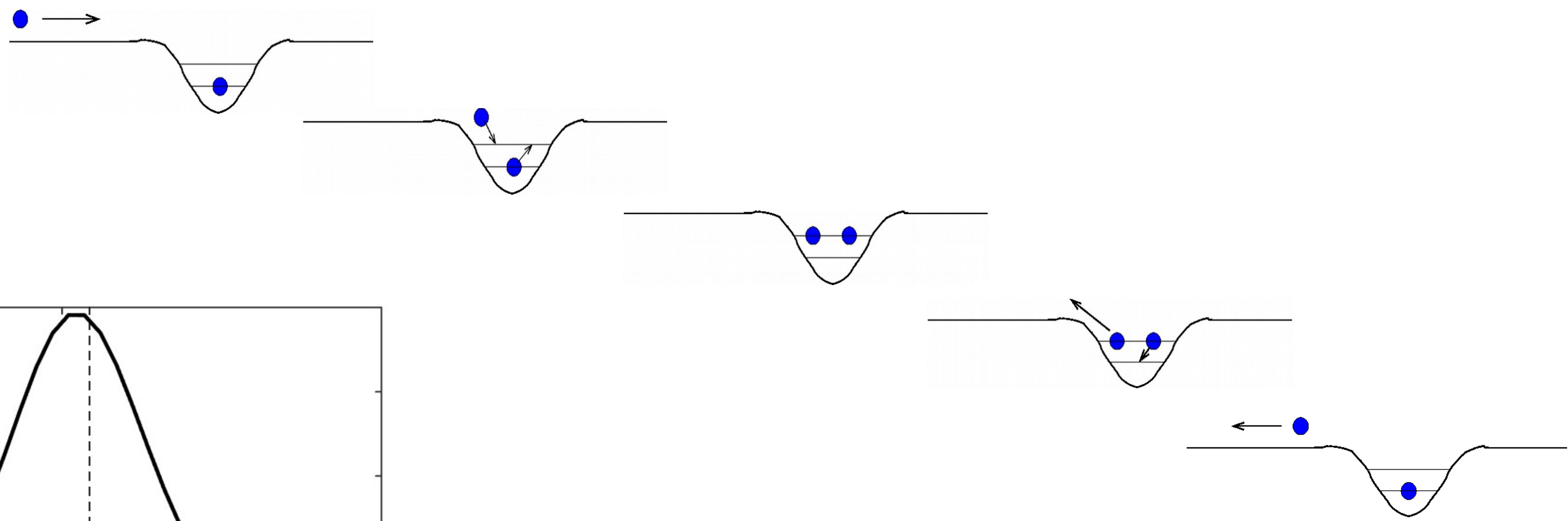
Why?

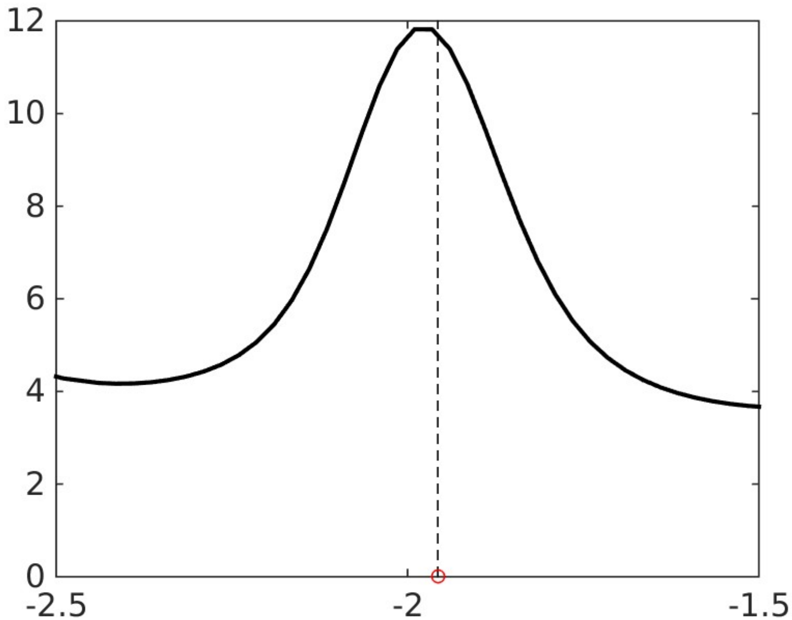


Photon Capture

Guess what: The resonance dominates

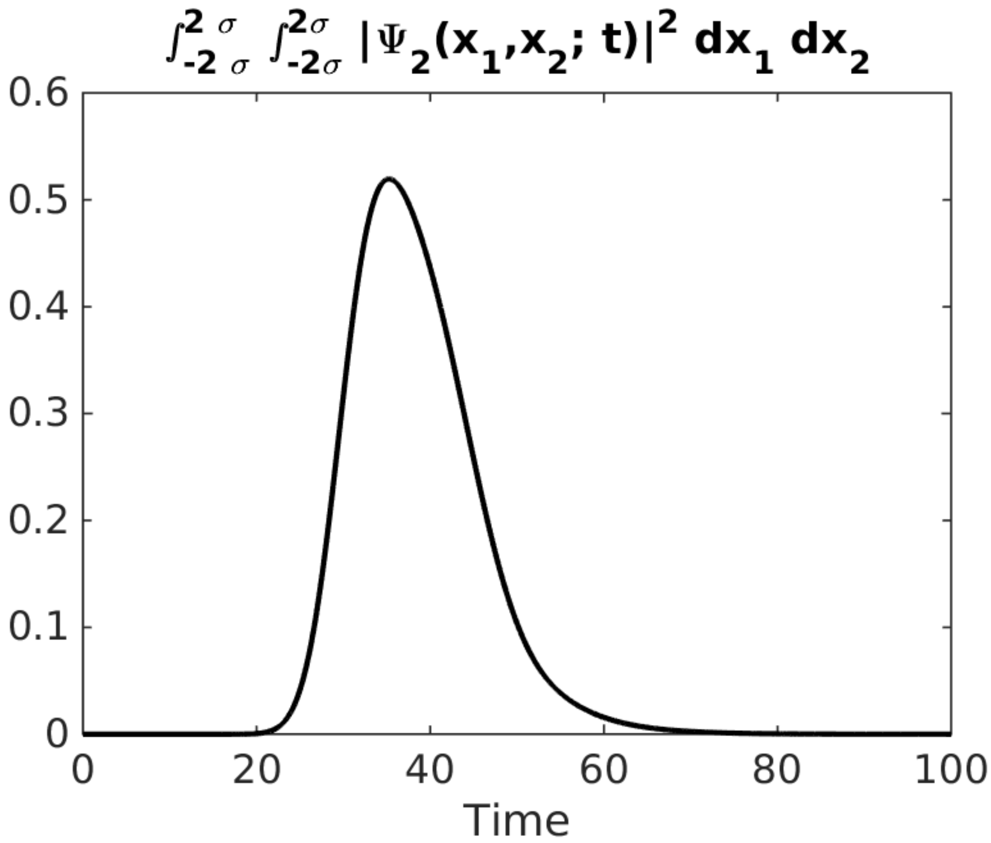
Why?



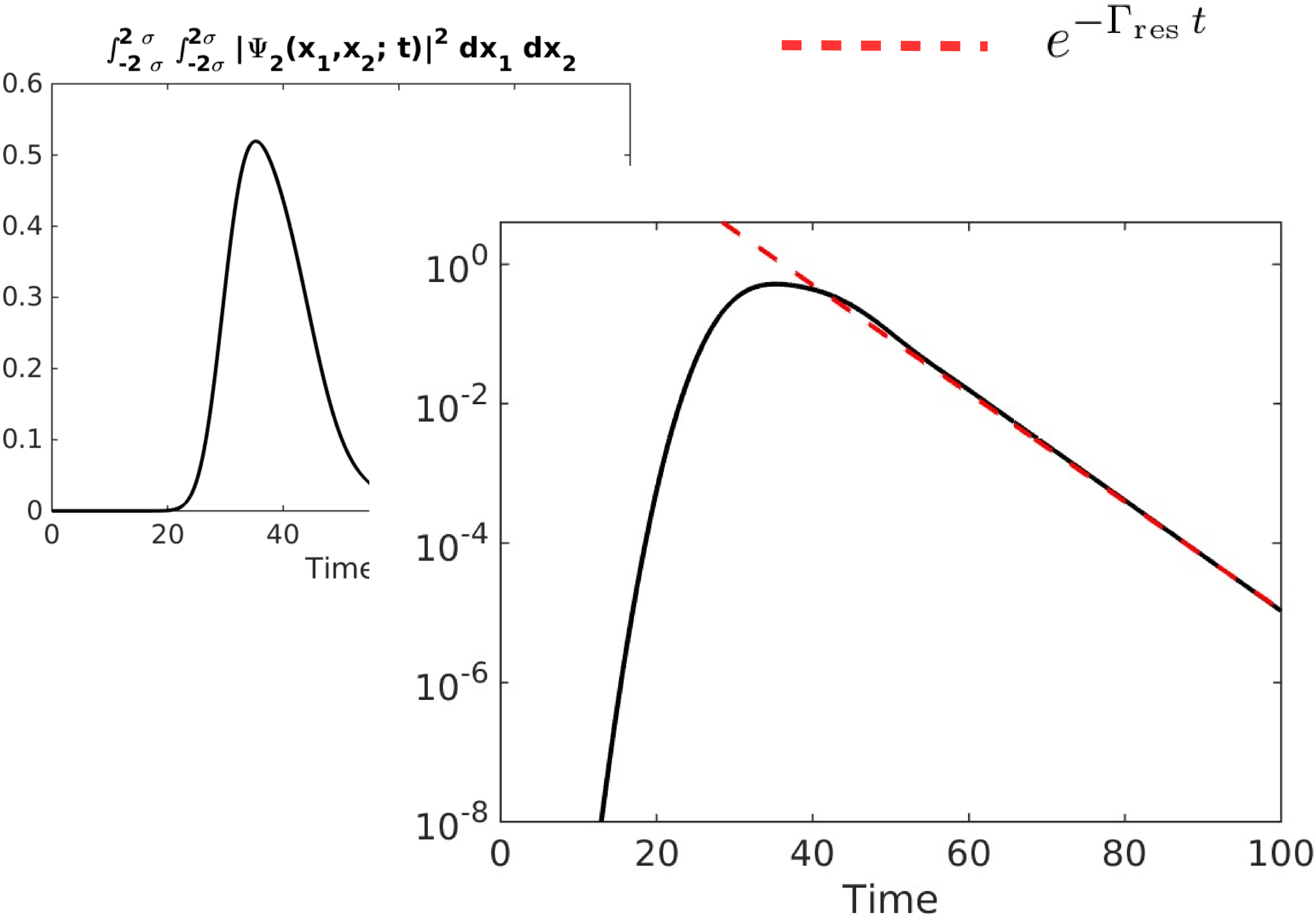


$$\int_{-\infty}^{\infty} \int_{-2\sigma}^{2\sigma} \int_{-2\sigma}^{2\sigma} \left|\Psi_2(x_1, x_2; t)\right|^2 \, dx_1 dx_2 \, dt$$

Exponential decay?



Exponential decay?



Capture: How?

Capture rate according to Fermi's golden rule:

$$\frac{\mathrm{d}}{\mathrm{d}t} P_b \approx 2\pi \sum_{\vec{k}} \left| \left\langle \Phi_b, 1_{\vec{k}} \right| H_I \left| \Psi_{\mathrm{init}}, 0 \right\rangle \right|^2 \delta\left(E_{\mathrm{init}} - (\varepsilon_b + \omega(k)) \right)$$

Capture: How?

Capture rate according to Fermi's golden rule:

$$\frac{\mathrm{d}}{\mathrm{d}t}P_b \approx 2\pi \sum_{\vec{k}} \left| \langle \Phi_b, 1_{\vec{k}} \right| H_I \left| \Psi_{\text{init}}, 0 \rangle \right|^2 \delta\left(E_{\text{init}} - (\varepsilon_b + \omega(k))\right)$$

Slightly modified:

$$\frac{\mathrm{d}}{\mathrm{d}t}P_b(t) \approx 2\pi \sum_{\vec{k}} \left| \langle \Phi_b, 1_{\vec{k}} \right| H_I \left| \Psi(t), 0 \rangle \right|^2 \delta\left(\langle E \rangle - (\varepsilon_b + \omega(k))\right)$$

Probability:

$$P_{\text{capture}} = \sum_b \int_0^\infty \dot{P}_b(t)\,\mathrm{d}t$$

Capture: How?

Capture rate according to Fermi's golden rule:

$$\frac{\mathrm{d}}{\mathrm{d}t} P_b \approx 2\pi \sum_{\vec{k}} \left| \langle \Phi_b, 1_{\vec{k}} \right| H_I \left| \Psi_{\mathrm{init}}, 0 \rangle \right|^2 \delta \left(E_{\mathrm{init}} - (\varepsilon_b + \omega(k)) \right)$$

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Probability:

$$P_{\mathrm{capture}} = \sum_b \int_0^\infty \dot{P}_b(t)\, \mathrm{d}t$$

Bound states:
Imaginary time; $t \to -it$

Capture: How?

Photons or phonons?

$$H_I^{\text{photon}} = i\vec{r} \cdot \sum_{\vec{k}} \sqrt{\frac{2\pi}{\omega(k)V}} \left(\vec{u}_{\vec{k}} \hat{a}_{\vec{k}} e^{\vec{k}\cdot\vec{r}} - \vec{u}_{\vec{k}} \hat{a}^{\dagger}_{\vec{k}} e^{-\vec{k}\cdot\vec{r}} \right)$$

$$H_I^{\text{phonon}} \propto \sum_{\vec{k}} \frac{k}{\sqrt{\omega(k)}} \left(\hat{a}_{\vec{k}} e^{i\vec{k}\cdot\vec{r}} + \hat{a}^{\dagger}_{\vec{k}} e^{-i\vec{k}\cdot\vec{r}} \right)$$

Capture: How?

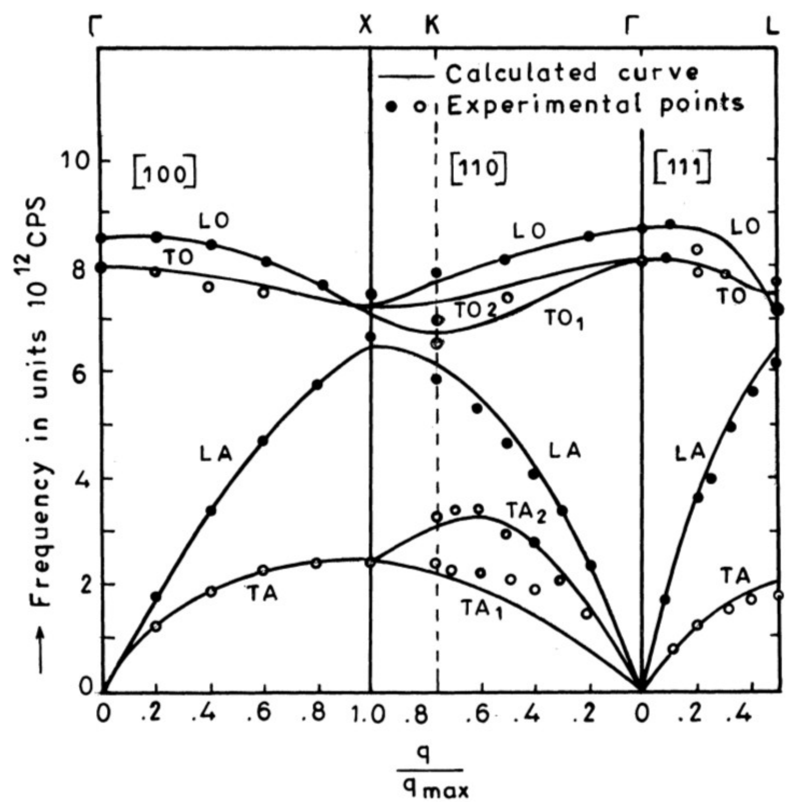
Dispersion relations

Photons or phonons?

$$k = \frac{\omega}{c}$$

$$H_I^{\text{photon}} = i\vec{r}\cdot\sum_{\vec{k}}\sqrt{\frac{2\pi}{\omega(k)V}}\left(\vec{u}_{\vec{k}}\hat{a}_{\vec{k}}e^{\vec{k}\cdot\vec{r}} - \vec{u}_{\vec{k}}\hat{a}^{\dagger}_{\vec{k}}e^{-\vec{k}\cdot\vec{r}}\right)$$

$$H_I^{\text{phonon}} \propto \sum_{\vec{k}}\frac{k}{\sqrt{\omega(k)}}\left(\hat{a}_{\vec{k}}e^{i\vec{k}\cdot\vec{r}} + \hat{a}^{\dagger}_{\vec{k}}e^{-i\vec{k}\cdot\vec{r}}\right)$$



R. K. Ram, S. S. Kushwaha, *J. Phys. Soc. Japan* **54**, 617 (1985)

Capture: How?

Photons or phonons?

$$H_I^{\text{photon}} = i\vec{r}\cdot\sum_{\vec{k}}\sqrt{\frac{2\pi}{\omega(k)V}}\left(\vec{u}_{\vec{k}}\hat{a}_{\vec{k}}e^{\vec{k}\cdot\vec{r}} - \vec{u}_{\vec{k}}\hat{a}^{\dagger}_{\vec{k}}e^{-\vec{k}\cdot\vec{r}}\right)$$

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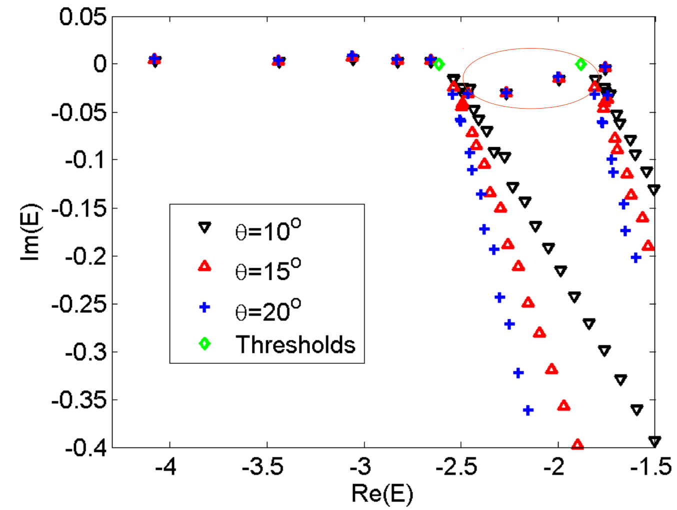
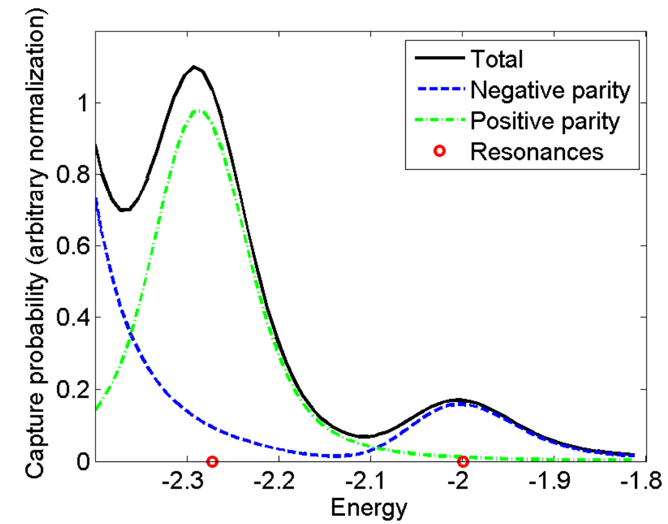
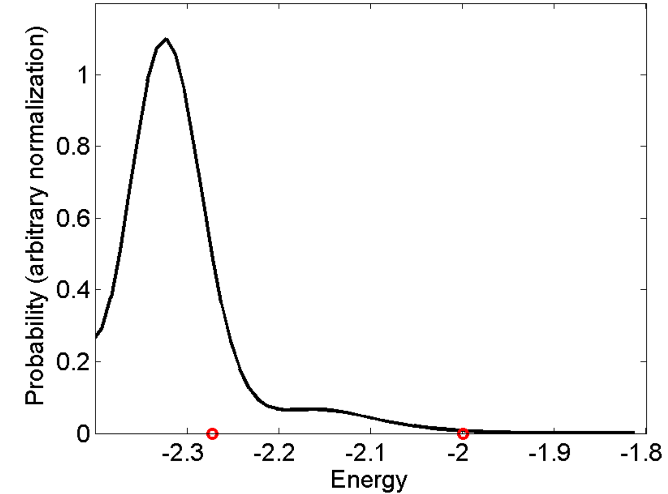
a)



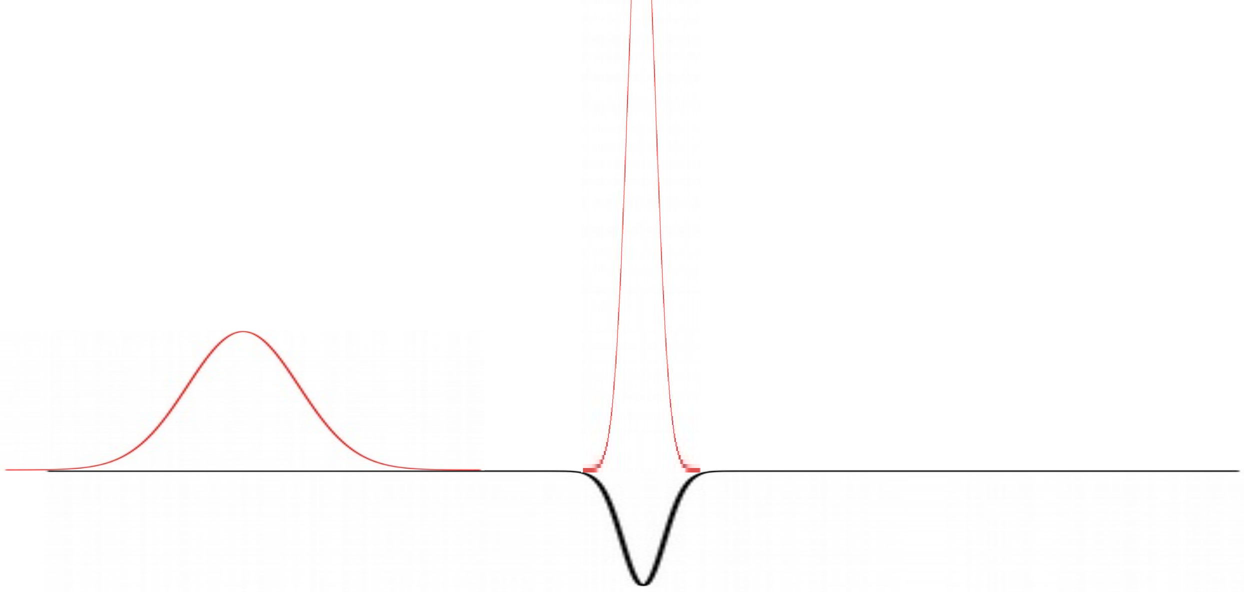
b)



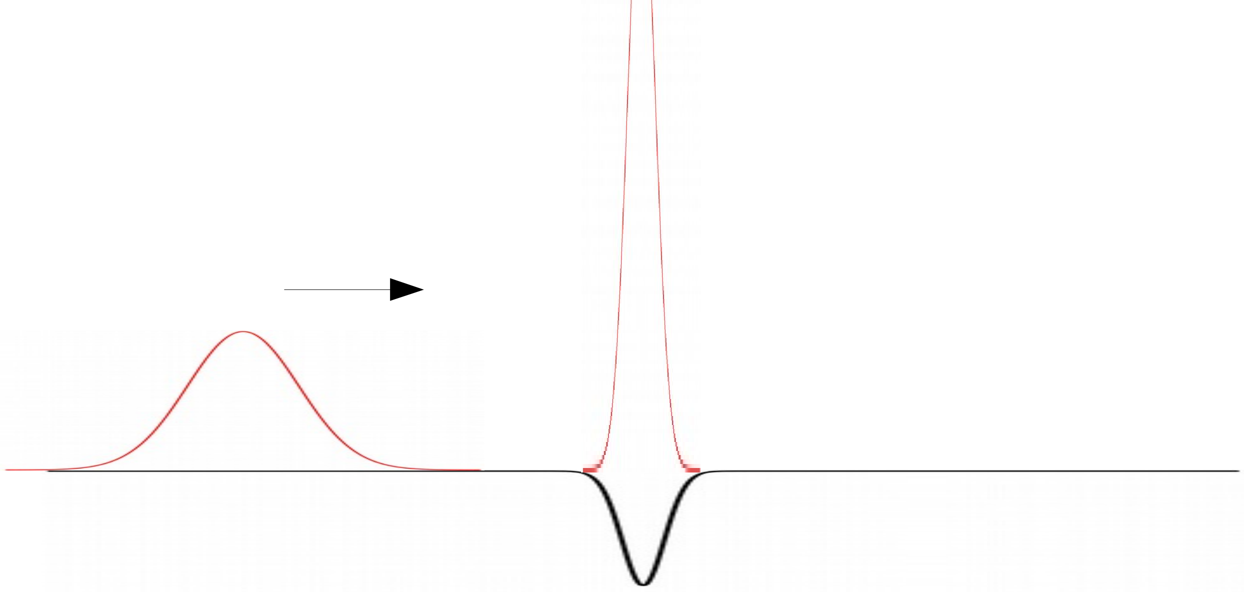
c)

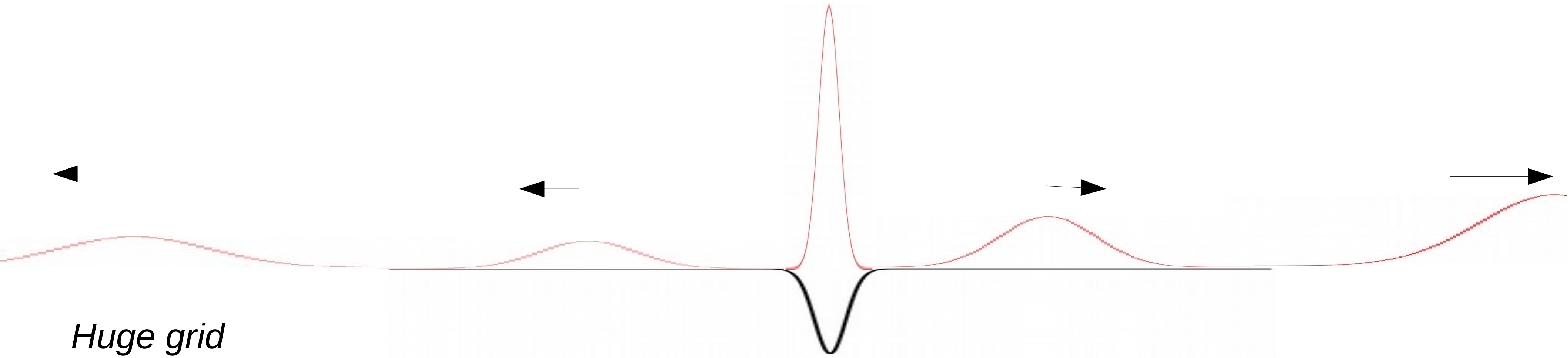


Problem



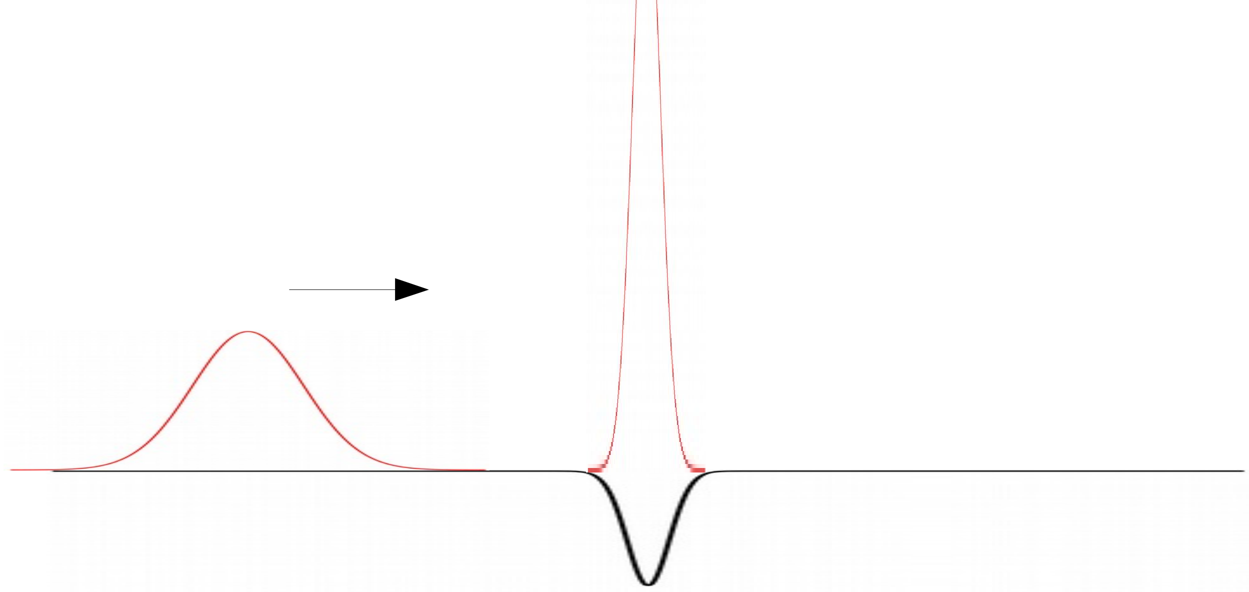
Problem

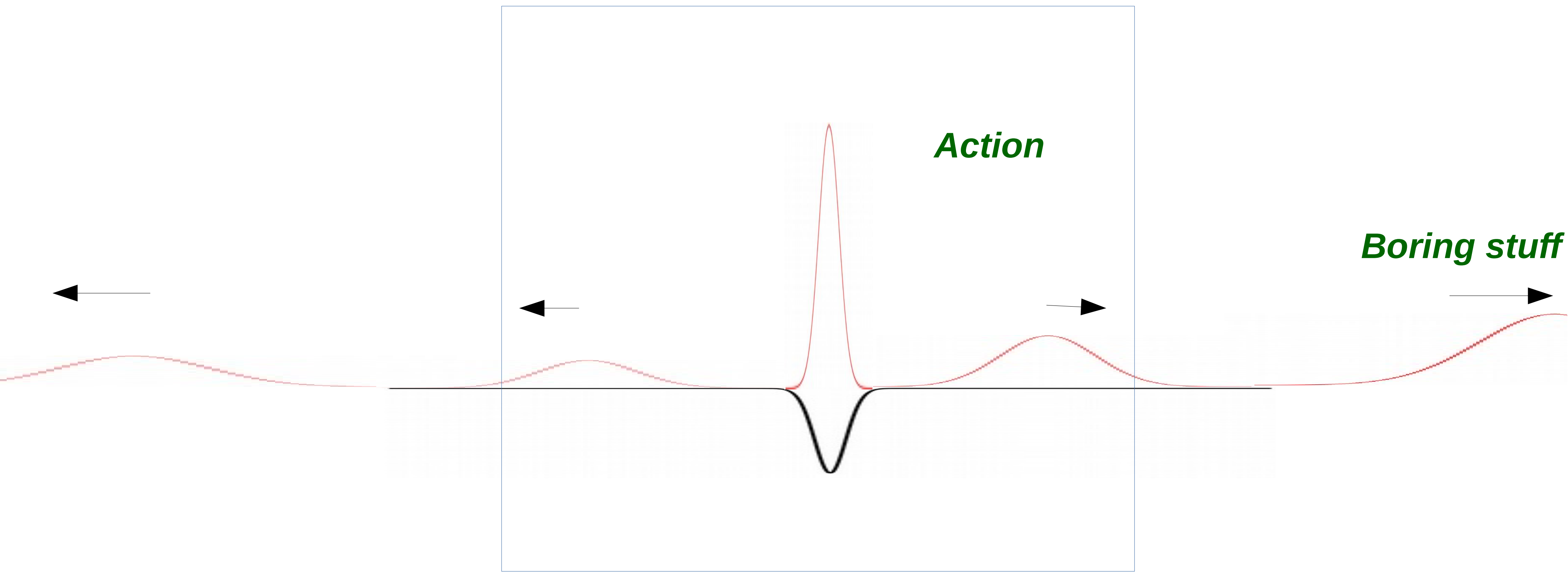




Huge grid

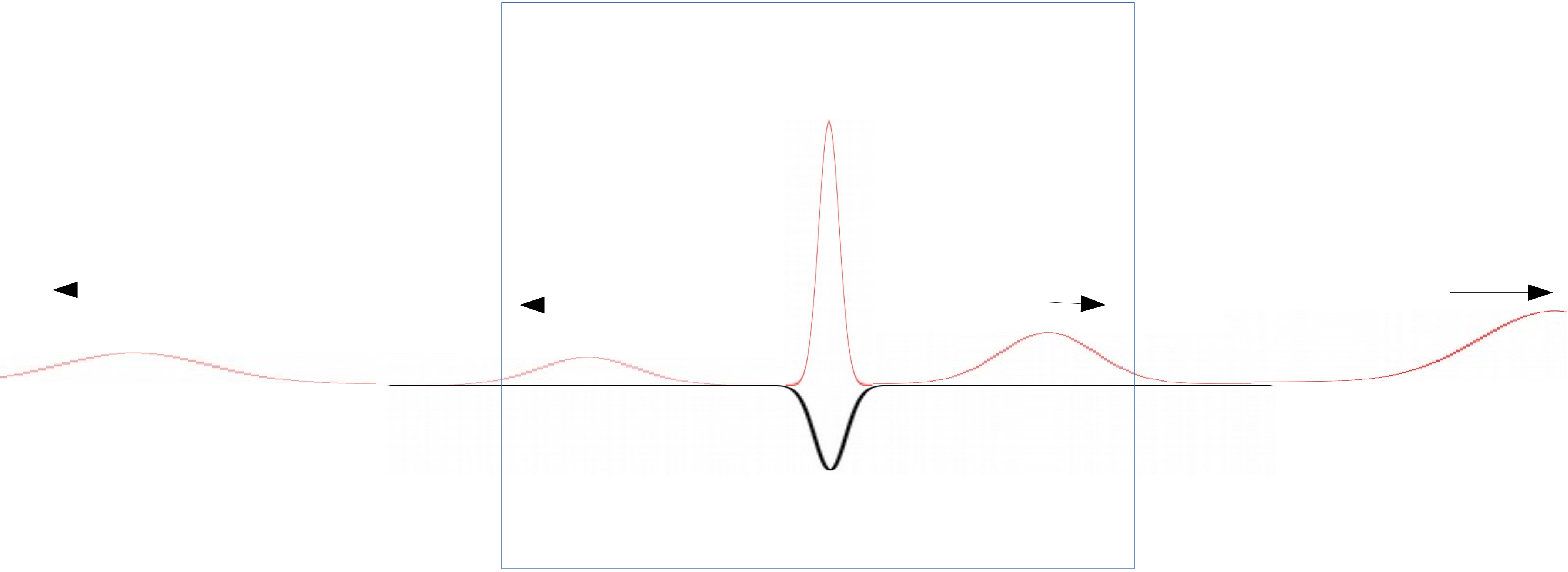
Problem





Problem

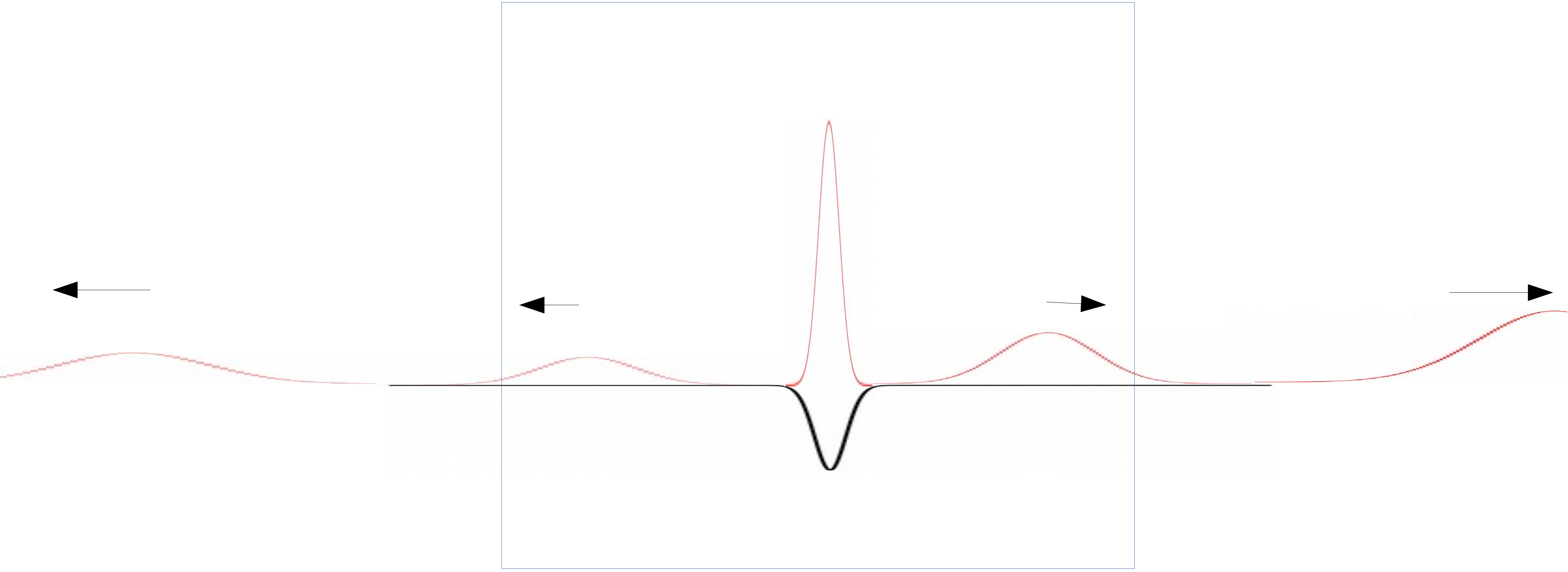
Complex absorbing potential? $V(x) \rightarrow V(x) - i\Gamma(x)$



Problem

Complex absorbing potential? $V(x) \to V(x) - i\Gamma(x)$

Eventually: $\Psi_2(x_1, x_2; t) \equiv 0$

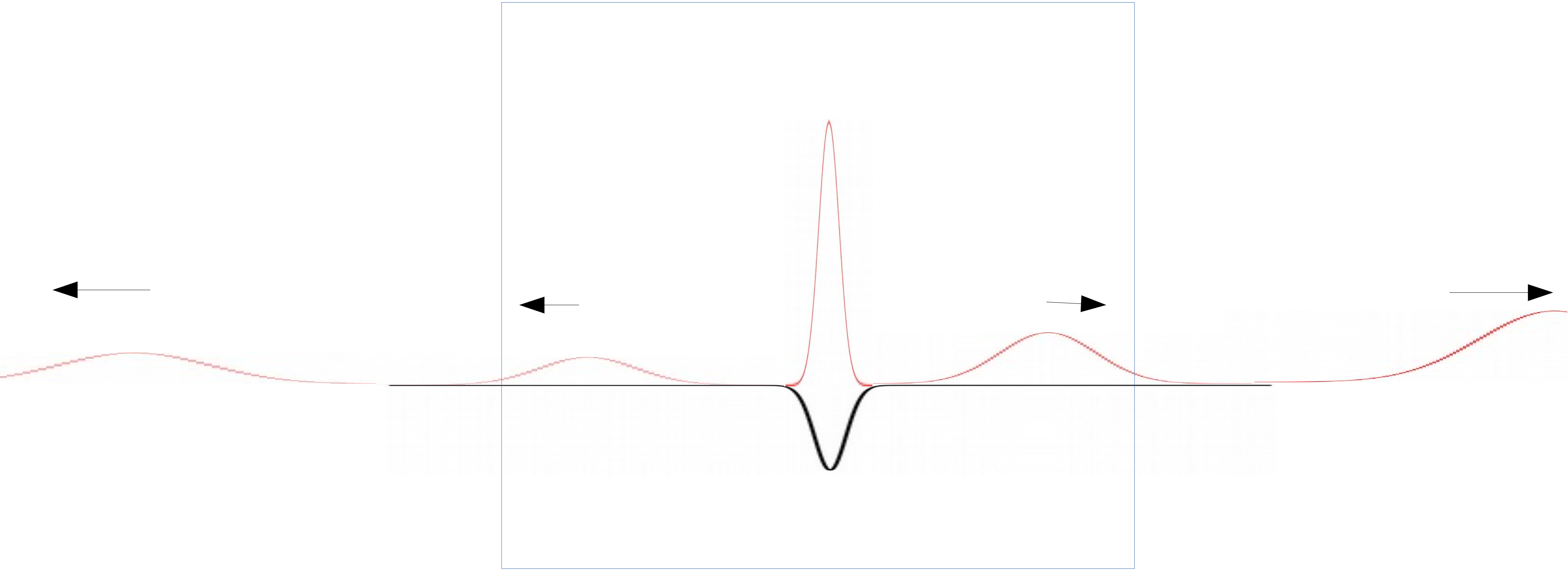


Problem

Complex absorbing potential? $V(x) \rightarrow V(x) - i\Gamma(x)$

Eventually: $\Psi_2(x_1, x_2; t) \equiv 0$

Feed a 1-particle Schrödinger equation?

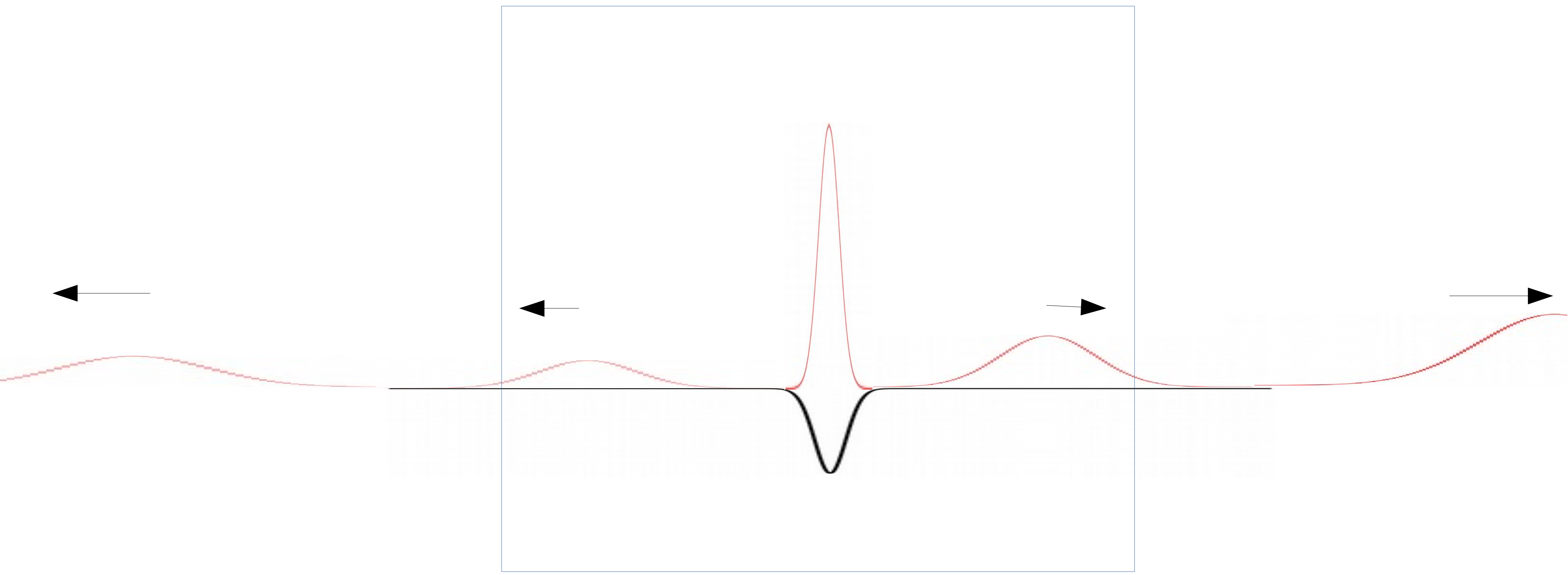


Problem

Complex absorbing potential? $V(x) \rightarrow V(x) - i\Gamma(x)$

Eventually: $\Psi_2(x_1, x_2; t) \equiv 0$

Feed a 1-particle Schrödinger equation? **NO**



Solution: The Lindblad equation

$$i\frac{\mathrm{d}}{\mathrm{d}t}\rho = [H,\rho] - \sum_{k,l}\gamma_{k,l}\left(\left\{a_k^\dagger a_l,\rho\right\} - 2a_l\rho a_k^\dagger\right)$$

Markovian
Preserve trace and positivity manifestly

G. Lindblad, *Commun. Math. Phys.* **48**, 119 (1976)

V. Gorini, A. Kossakowski, E. Sudarshan, *J. Math. Phys.* **17**, 821 (1976)

Solution: The Lindblad equation

$$i\frac{\mathrm{d}}{\mathrm{d}t}\rho = [H,\rho] - \sum_{k,l}\gamma_{k,l}\left(\left\{a_k^\dagger a_l,\rho\right\} - 2a_l\rho a_k^\dagger\right) = H_{\mathrm{eff}}\rho - \rho H_{\mathrm{eff}}^\dagger + 2\sum_{k,l}\gamma_{k.l}a_l\rho a_k^\dagger$$

$$H_{\mathrm{eff}} = H - i\hat{\Gamma}$$

$$\hat{\Gamma} = \sum_{k}\Gamma(x_k)c_k^\dagger c_k = \sum_{k,l}\gamma_{k,l}a_k^\dagger a_l$$

$$\gamma_{k,l} = \Gamma(x_k)\delta_{k,l}$$

$$a_k = c_k$$

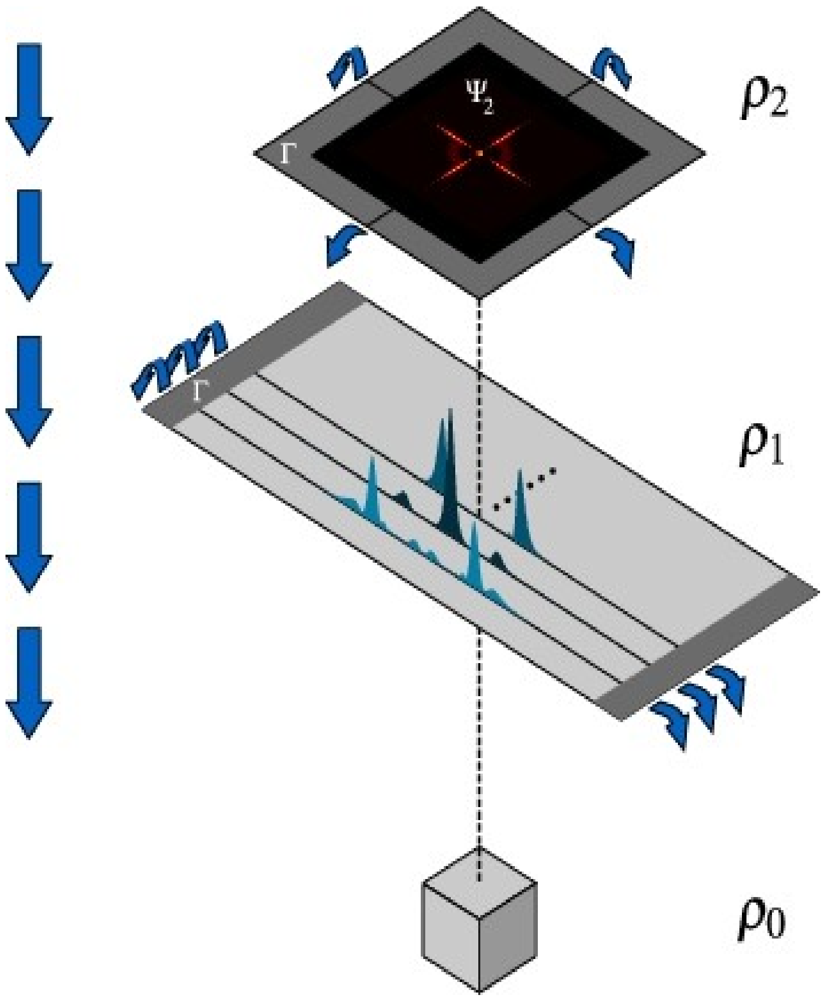
Source term

$$\mathcal{S}[\rho_N] \equiv 2i\sum_k \Gamma(x_k)c_k\rho_N c_k^\dagger$$

S. S., S. Kvaal, *J. Phys. B* **43**, 065004 (2010)

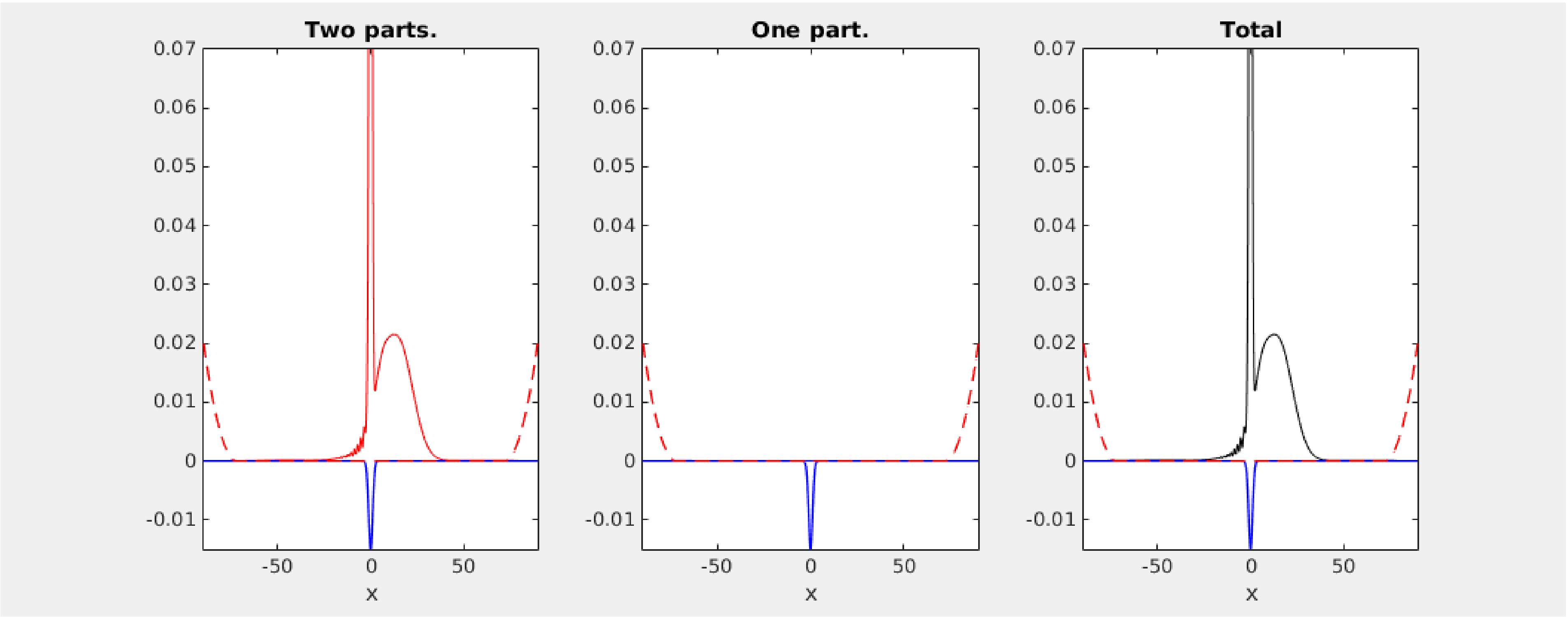
Solution: The Lindblad equation

$$i\frac{\mathrm{d}}{\mathrm{d}t}\rho = [H,\rho] - \sum_{k,l}\gamma_{k,l}\left(\left\{a_k^\dagger a_l,\rho\right\} - 2a_l\rho a_k^\dagger\right) = H_{\text{eff}}\rho - \rho H_{\text{eff}}^\dagger + 2\sum_{k,l}\gamma_{k.l}a_l\rho a_k^\dagger$$

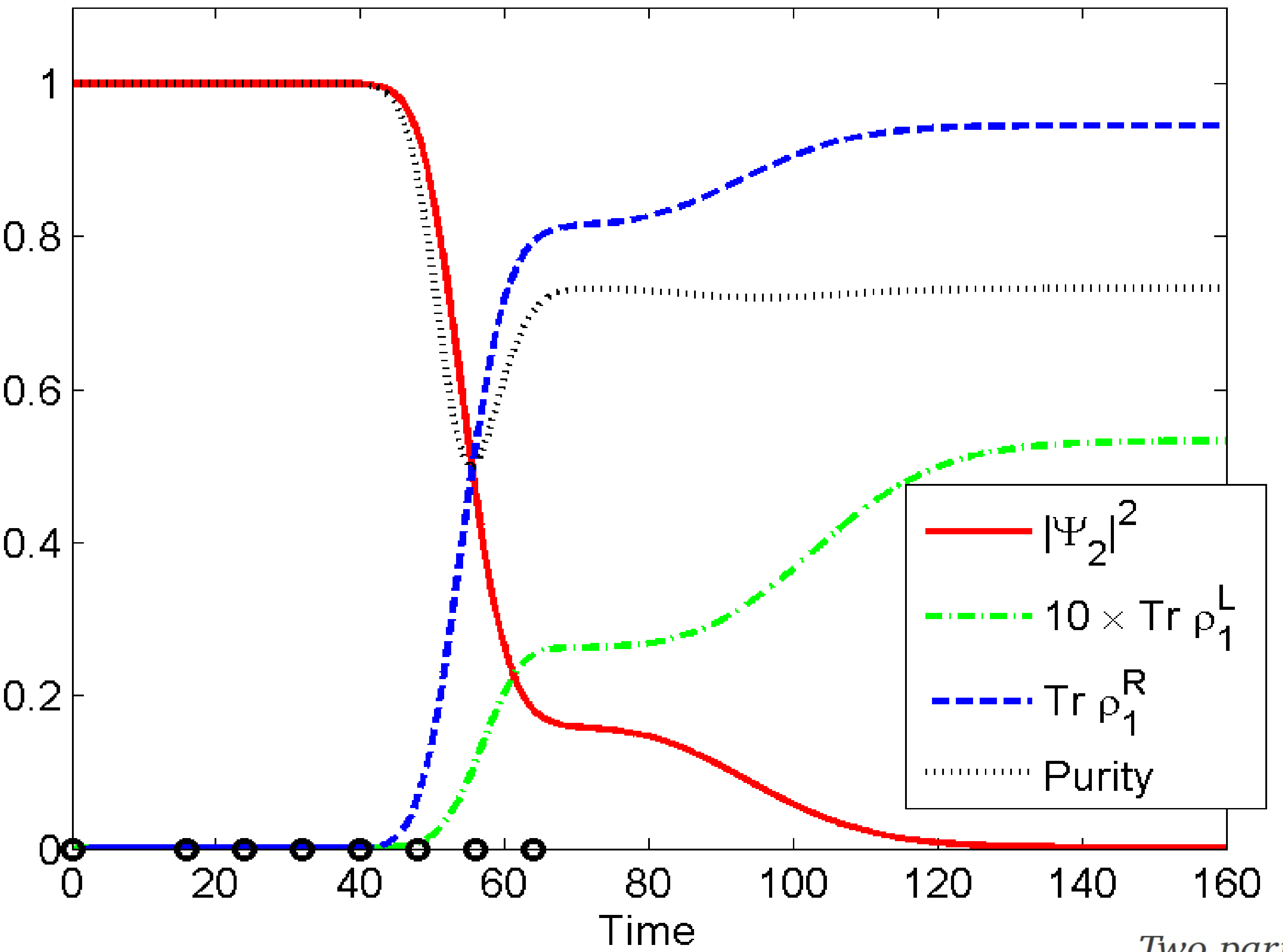


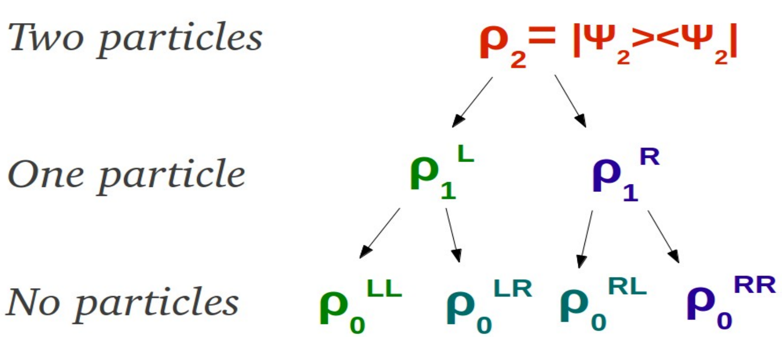
$$\begin{aligned}
\rho_2 &= |\Psi_2\rangle\langle\Psi_2|, \quad i\dot{\Psi}_2 = H_{\text{eff},2}\Psi_2 \\
i\dot{\rho}_1 &= H_{\text{eff},1}\rho_1 - \rho_1 H_{\text{eff},1}^\dagger + \mathcal{S}[\rho_2] \\
i\dot{\rho}_0 &= \mathcal{S}[\rho_1], \quad (\rho_0 = 1 - \mathrm{Tr}\rho_2 - \mathrm{Tr}\rho_1)
\end{aligned}$$

S. S., T. Birkeland, S. Kvaal, R. Nepstad, M. Førre, *J. Phys. B* **44**, 215003 (2011)

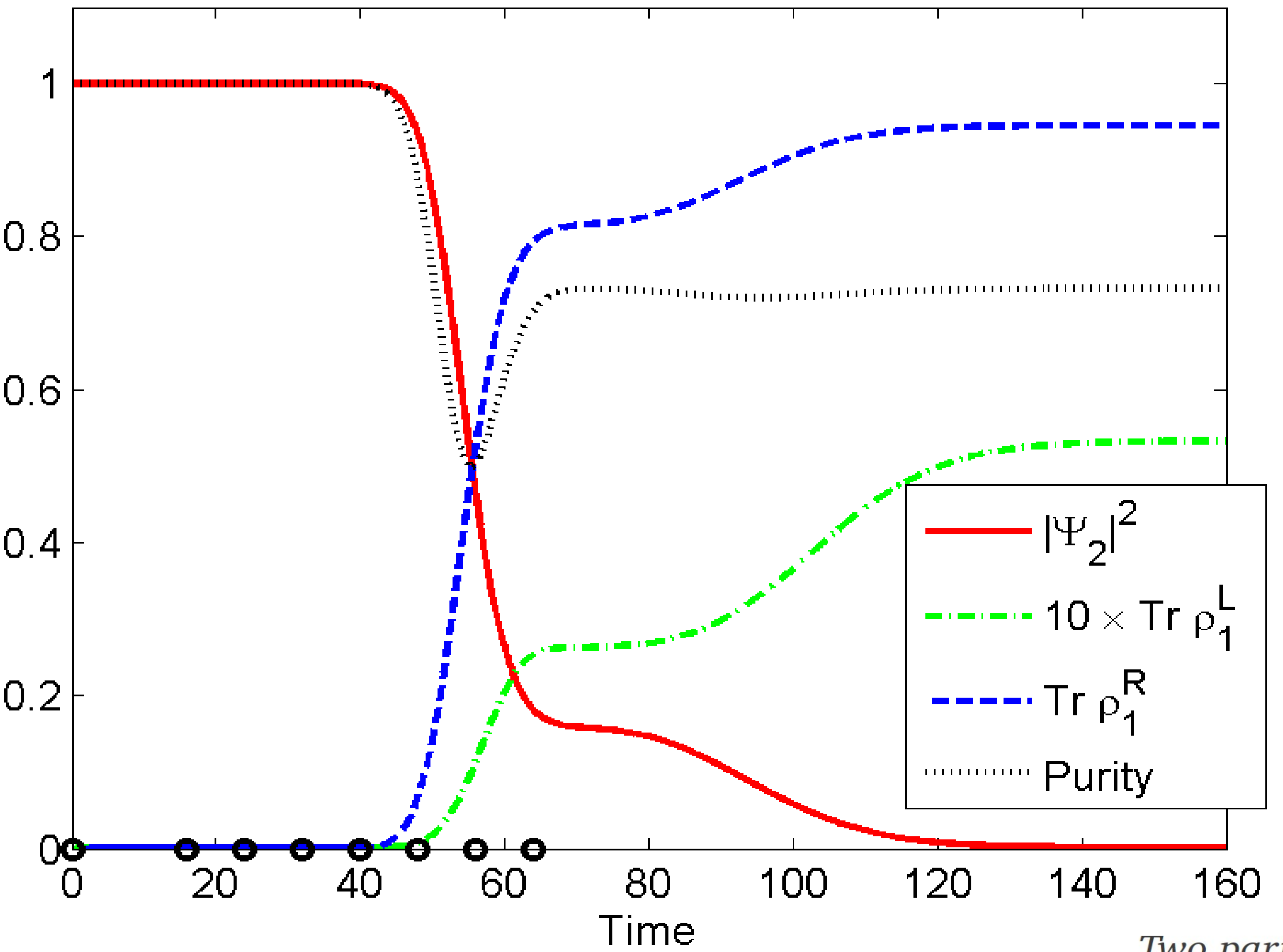

Two parts.
One part.
Total
0.07
0.06
0.05
0.04
0.03
0.02
0.01
0
-0.01
-50
0
50
x

How many particles are there on the grid?





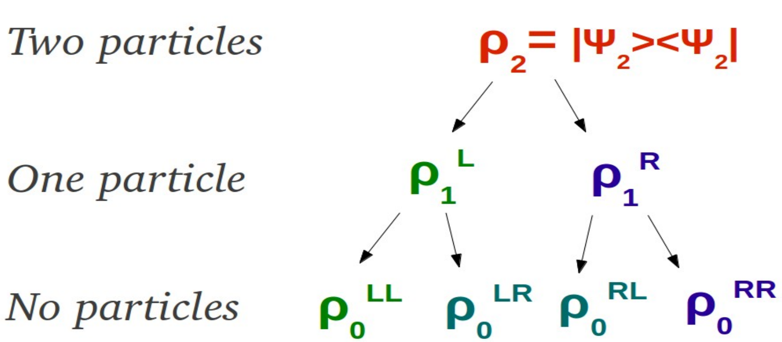
How many particles are there on the grid?



$$R = \lim_{t\to\infty} \mathrm{Tr}\,\rho_1^{\mathrm{L}}(t)$$

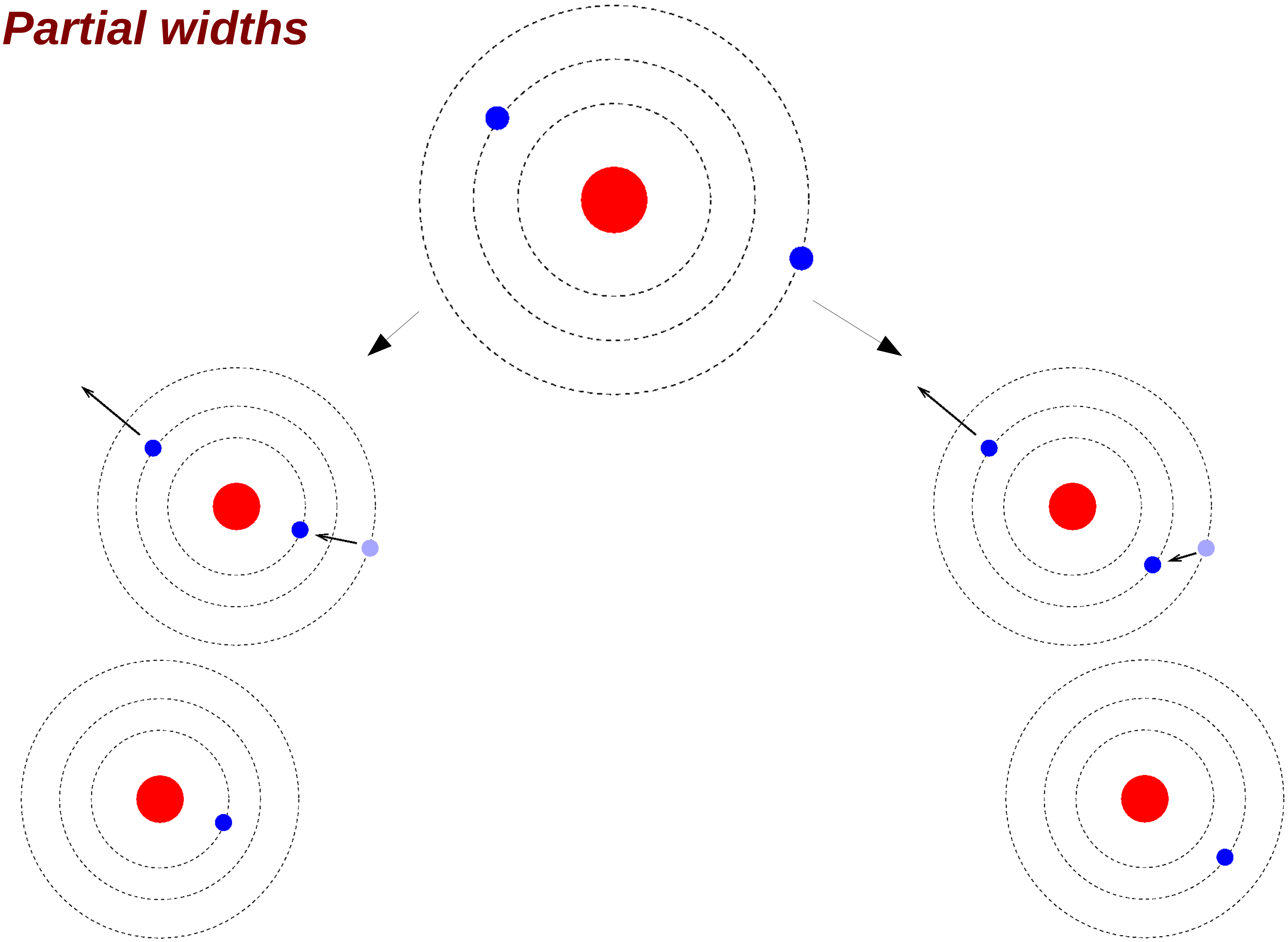
$$T = \lim_{t\to\infty} \mathrm{Tr}\,\rho_1^{\mathrm{R}}(t)$$

$$P^{(\mathrm{b})}_{\mathrm{excite}}(t) = \langle \varphi_{\mathrm{b}} | \rho_1(t) | \varphi_{\mathrm{b}} \rangle$$

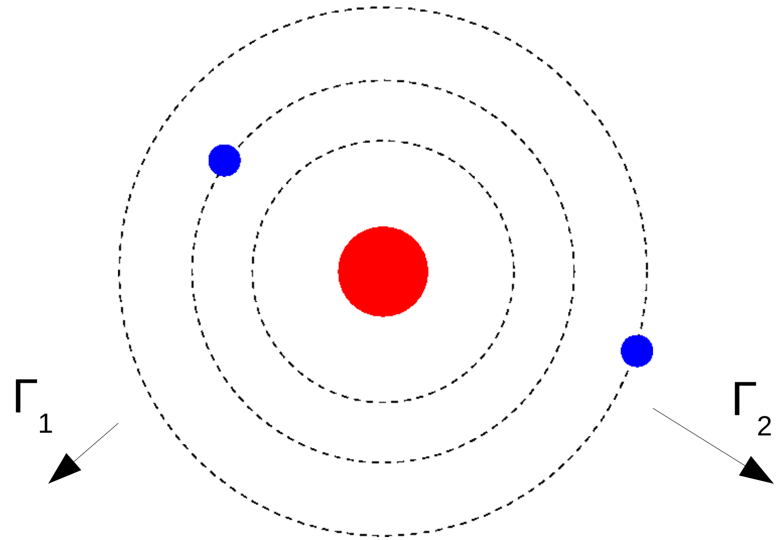


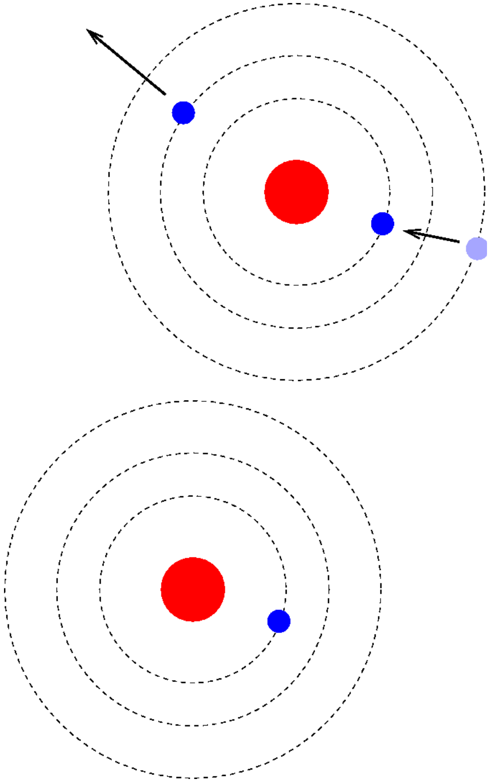
Another application of the same formalism:

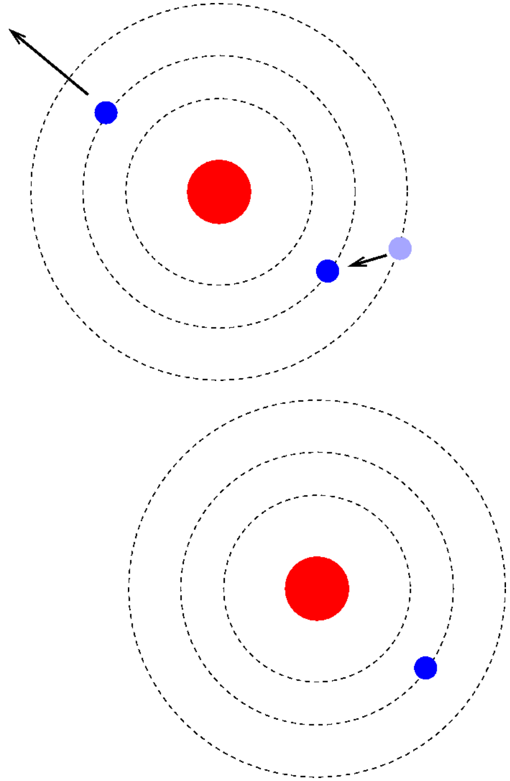
Partial widths



Expected classically:

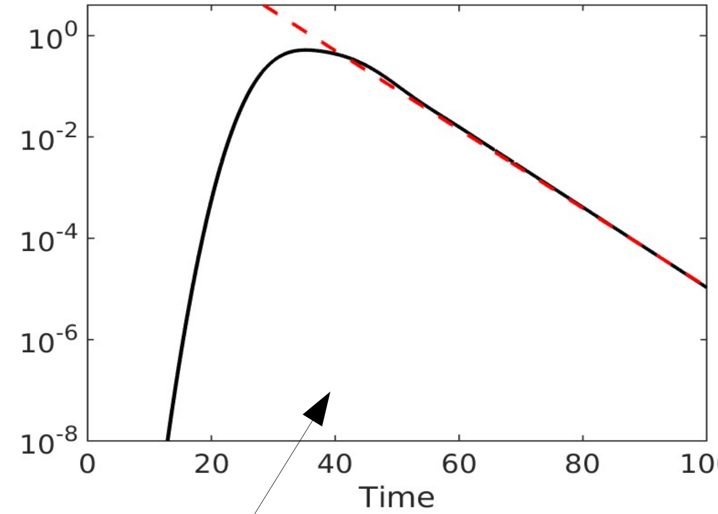


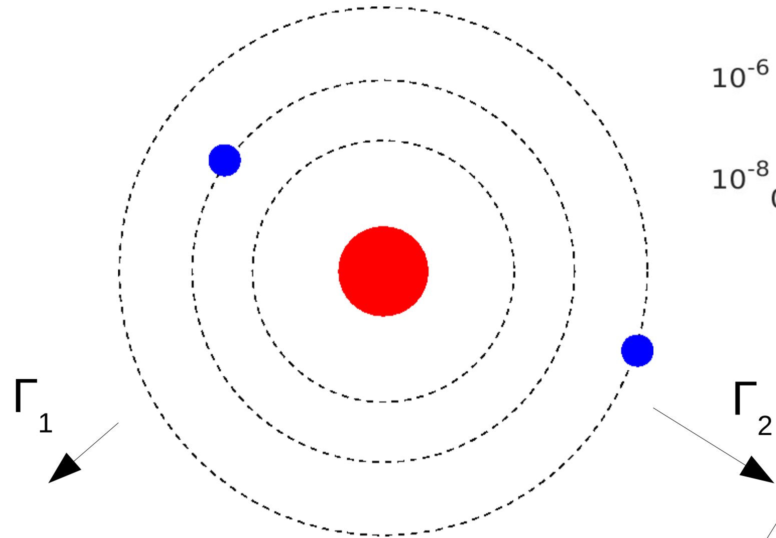


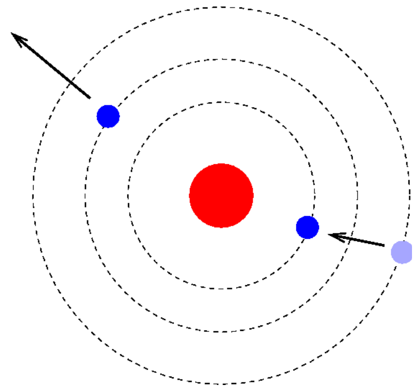


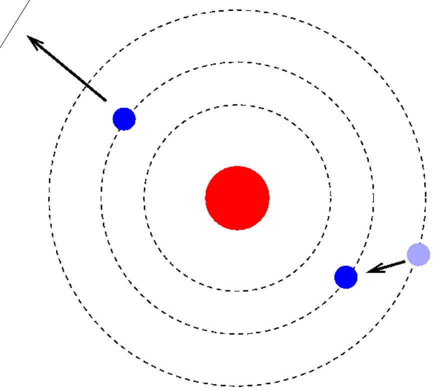
$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t} P_{\mathrm{res}} &= -\Gamma_{\mathrm{res}} P_{\mathrm{res}} \\
\frac{\mathrm{d}}{\mathrm{d}t} P_{b} &= \Gamma_{b} P_{\mathrm{res}} \\
\sum_{b} \Gamma_{b} &= \Gamma_{\mathrm{res}}
\end{aligned}
$$

Expected classically:





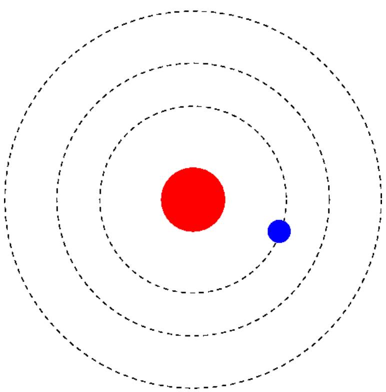


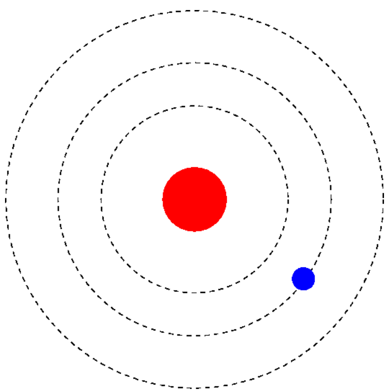


$$\frac{\mathrm{d}}{\mathrm{d}t}P_{\mathrm{res}} = -\Gamma_{\mathrm{res}}P_{\mathrm{res}}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}P_{b} = \Gamma_{b}P_{\mathrm{res}}$$

$$\sum_{b}\Gamma_{b} = \Gamma_{\mathrm{res}}$$





The Lindblad equation gives

$$\begin{aligned} i\dot{\Psi}_N &= H_{\rm eff}\Psi_N \\ i\dot{\rho}_{N-1} &= [H_{\rm eff}, \rho_{N-1}] + 2\sum_k \Gamma(x_k) c_k |\Psi_2\rangle\langle\Psi_2| c_k^\dagger \end{aligned}$$

which, in turn, gives

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t} P_{\rm res} &= -\Gamma_{\rm res} P_{\rm res} \\ \frac{\mathrm{d}}{\mathrm{d}t} P_b &= \Gamma_b P_{\rm res} \end{aligned}$$

provided that

$$\begin{aligned} P_{\rm res} &= \left|\langle\Psi_{\rm res}|\Psi_N(t)\rangle\right|^2 \\ P_b &= \langle\Phi_b|\rho_{N-1}|\Phi_b\rangle \\ \Gamma_b &= \sum_k \Gamma(x_k)\left|\langle\Phi_b|c_k|\Psi_{\rm res}\rangle\right|^2 \end{aligned}$$

S. S., *Phys. Rev. A* **85**, 062518 (2012)

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$$\Psi_N(t) = e^{-i(E_{\text{res}} - i\Gamma_{\text{res}}/2)t}\Psi_{\text{res}}, \quad |\Psi_N(t)|^2 = e^{-\Gamma_{\text{res}} t}$$

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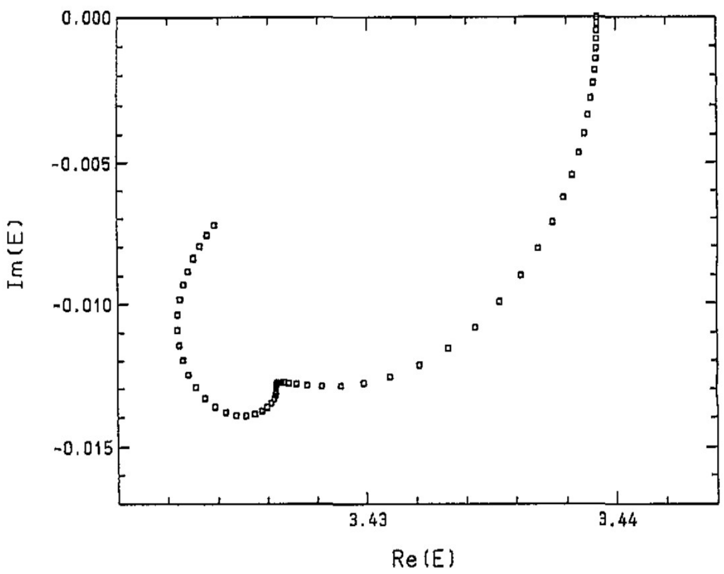
Trace conservation => $\sum_b \Gamma_b = \Gamma_{\text{res}}$

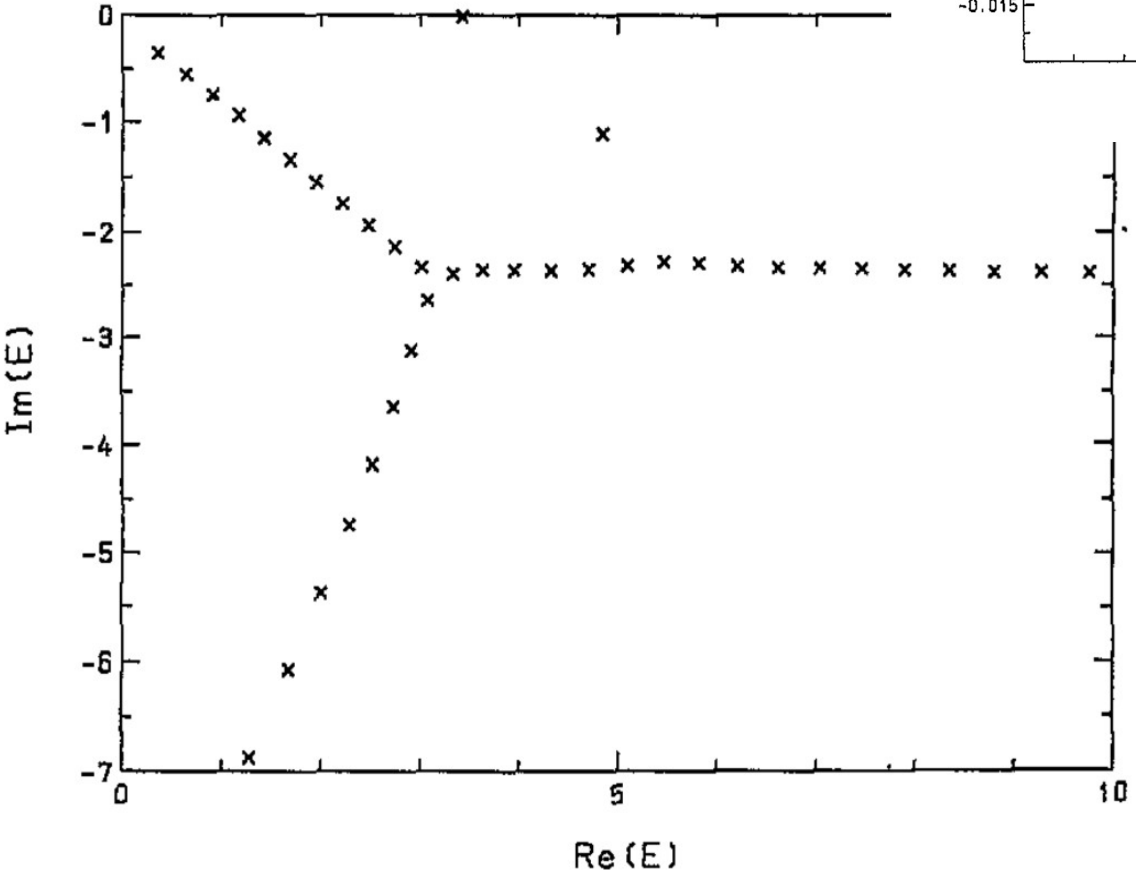
S. S., *Phys. Rev. A* **85**, 062518 (2012)

-*Can* use CAP-s in order to identify resonances

U. V. Riss, H.-D. Meyer, J. Phys. B **26**, 4503 (1993)

R Santra, *Phys. Rev. A* **74**, 034701 (2012)





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R Santra, *Phys. Rev. A* **74**, 034701 (2012)

Better(?): Exterior complex scaling

$$r \to R(r) = \begin{cases} r, & r \leq R_0 \\ R_0 + (r - R_0)e^{i\theta}, & r > R_0 \end{cases}$$

$$\begin{aligned} h^{\mathrm{I}}_{k,l} &= \int\limits_{r>R_0} d^3\mathbf{r}\ (\chi_k(\mathbf{r}))^* \left[-\sin(2\theta)\frac{\hbar^2}{2m}\nabla^2 + \mathrm{Im}\, V_1(R(\mathbf{r}))\right] \chi_l(\mathbf{r}) \quad \text{and} \\ V^{\mathrm{I}}_{pq,rs} &= \int\limits_{r>R_0}\int\limits_{r'>R_0} d^3\mathbf{r}\, d^3\mathbf{r}'\ (\chi_p(\mathbf{r}))^* (\chi_q(\mathbf{r}'))^* \left[\mathrm{Im}\, V_2(R(\mathbf{r}), R(\mathbf{r}'))\right] \chi_r(\mathbf{r})\chi_s(\mathbf{r}') \end{aligned}$$

$$H^{\mathrm{ah}} = \sum_{kl} h^{\mathrm{I}}_{k,l} c^\dagger_k c_l + \frac{1}{2}\sum_{pqrs} V^{\mathrm{I}}_{pq,rs} c^\dagger_p c^\dagger_q c_s c_r$$

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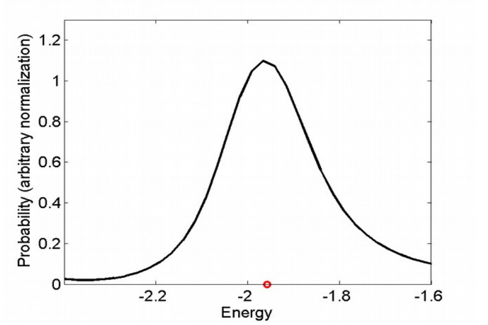
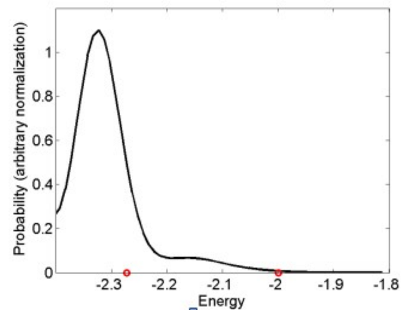
$$\Gamma_b = 2\sum_{k,l} h^{\mathrm{I}}_{k,l} \left\langle \Phi_b^{(N-1)} \middle| c_l \middle| \Psi^{(N)}_{\mathrm{res}} \right\rangle \left\langle \Psi^{(N)}_{\mathrm{res}} \middle| c_k^\dagger \middle| \Phi_b^{(N-1)} \right\rangle$$

Brief summery

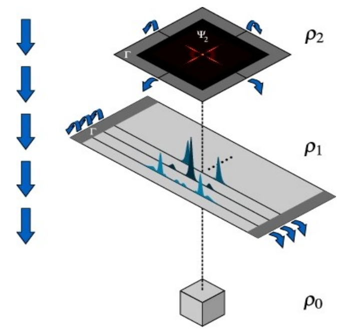
- Resonances do indeed play a crucial role – both in scattering and capture in a quantum dot



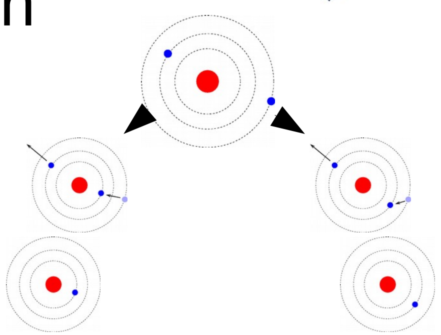
- Facilitates putting electrons into quantum dots?



- However: For phonon capture this dependence tends to be washed out by strong energy dependence in the interaction.



- Methodwise:
  Absorbing boundaries -> the Lindblad equation



- Application of the same formalism:
  Compact expressions for partial widths


Grattis!