Linear response theory for open two-terminal quantum systems

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Stationary system

- Second quantization for open systems
- Statistical operator for the unperturbed system
- Expectation values; Landauer-Büttiker formalism

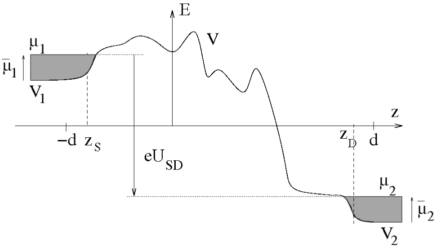
Small harmonic perturbation

- Linear response theory and RPA
- Effective potential inside the scattering region
- ac quantum admittance

Example: quantum capacitor

- Small-signal equivalent RC circuit
- Numerical results for MIS-type nanostructure

Conclusions



- planar structure; effective 1D system

$$\varphi(\boldsymbol{r}) = \psi^{(s)}(\epsilon, z)\frac{\exp\left(i\boldsymbol{k}_\perp \boldsymbol{r}_\perp\right)}{\sqrt{A}}$$

- reservoirs: $V(z < -d < z_S) = V_1$; $V(z > d > z_D) = V_2$
- scattering wave functions: $\psi^{(s)}(\epsilon, z)$, $s = 1, 2$

$$\left[-\frac{\hbar^2}{2m^*}\frac{d^2}{dz^2} + V(z) - \epsilon\right]\psi^{(s)}(\epsilon, z) = 0, \qquad z \in (-\infty, \infty)$$

- Hartree approximation $V(z) = V_{het}(z) + V_{el}(z)$
- we compute $\psi^{(s)}(\epsilon, z)$, over whole z axis, using the R-matrix formalism

- $\{\psi^{(s)}(\epsilon, z)\}$ form a continuos orthonormal basis
- introduce a k-space discretization: $k_j = j * \Delta k,\ j \in N$

$$\epsilon_1(k_j) = V_1 + \frac{\hbar^2 k_j^2}{2m^*} = V_2 + \frac{\hbar^2 k_j'^2}{2m^*}$$
$$\epsilon_2(k_j) = V_2 + \frac{\hbar^2 k_j^2}{2m^*} = V_1 + \frac{\hbar^2 k_j'^2}{2m^*}$$

- $\psi^{(s)}(\epsilon, z) \to \sqrt{\Delta k}\psi^{(s)}(\epsilon_s(k_j), z) = \psi_{sj}(z)$
- discrete orthonormal basis $\{\phi_\nu(\boldsymbol{r}_\perp)\}$ for $\boldsymbol{r}_\perp$ directions

$$\Downarrow$$

- discrete basis $\alpha \equiv (sj\nu)$

$$\varphi_\alpha(\boldsymbol{r}) = <\boldsymbol{r}|\varphi_\alpha> = \psi_{sj}(z)\phi_\nu(\boldsymbol{r}_\perp)$$

- creation and annihilation operators $\hat{c}_\alpha^\dagger$ and $\hat{c}_\alpha$, and CAR

$\Rightarrow$ one can use now the particle number representation

The unperturbed system

$$\hat{H}_0 = \int d^3r\, \hat{\Psi}^\dagger(\boldsymbol{r}) \left[-\frac{\hbar^2}{2m^*}\Delta + V_\perp(\boldsymbol{r}_\perp) + V(z)\right] \hat{\Psi}(\boldsymbol{r}),$$
$$= \sum_\alpha E_\alpha \hat{c}_\alpha^\dagger \hat{c}_\alpha,$$

is the stationary system under applied bias U_{SD}.
Ansatz for the statistical operator of the unperturbed system

$$\hat{\rho}_0 = \frac{1}{Z_0} \exp[-\beta(\hat{H}_0 - \mu_1 \hat{N}_1 - \mu_2 \hat{N}_2)]$$

with particle number operators $\hat{N}_s = \sum_{j\nu} \hat{c}_{sj\nu}^\dagger \hat{c}_{sj\nu}$ and $\hat{N} = \hat{N}_1 + \hat{N}_2$
and with the grand canonical partition function

$$Z_0 = \mathrm{Tr}\{\exp[-\beta(\hat{H}_0 - \mu_1 \hat{N}_1 - \mu_2 \hat{N}_2]\}$$
$$= \prod_\alpha (1 + \exp(-\beta(E_\alpha - \mu_s)))$$

$\Rightarrow$ mean value of the one-particle number operator

$$\bar{n}_\alpha = \langle \hat{c}_\alpha^\dagger \hat{c}_\alpha \rangle = \mathrm{Tr}\{\hat{\rho}_0 \hat{c}_\alpha^\dagger \hat{c}_\alpha\} = f_{FD}(E_\alpha - \mu_s), \qquad \alpha \equiv (sj\nu)$$

This is the ansatz done by Büttiker, PRB 46, 12485, (1992)

$\Rightarrow$ mean value of the particle density operator

$$\begin{aligned} \rho(\boldsymbol{r}) &= 2\mathrm{Tr}\left\{\hat{\rho}_0\, \hat{\Psi}^\dagger(\boldsymbol{r})\hat{\Psi}(\boldsymbol{r})\right\} = 2\sum_\alpha f_{FD}(E_\alpha - \mu_s)|\varphi_\alpha(\boldsymbol{r})|^2 \\ &= 2\sum_{s\nu}\int_{V_s}^{\infty} d\epsilon\; g_s(\epsilon) f_{FD}(E_\perp^\nu + \epsilon - \mu_s)\,|\phi_\nu(\boldsymbol{r}_\perp)|^2 \left|\psi^{(s)}(\epsilon, z)\right|^2 \end{aligned}$$

is the same as in the Landauer-Büttiker formalism

- external applied bias

$$U_{SD}(t) = U_{SD} + \delta U e^{-i(\omega+i\eta)t}$$

- perturbation in potential energy $\delta V(\boldsymbol{r},t) = \delta V(\boldsymbol{r}) \exp\left[-i(\omega+i\eta)t\right]$
- time dependent Hamiltonian

$$\hat{H} = \hat{H}_0 + \int d^3r\, \hat{\rho}(\boldsymbol{r})\delta V(\boldsymbol{r},t)$$

Linear response theory

- response in the particle density $\delta\rho(\boldsymbol{r},t) = \delta\rho(\boldsymbol{r})\exp(-i\omega t)$

$$\delta\rho(\boldsymbol{r}) = \int d^3r'\, \Pi_0(\boldsymbol{r},\boldsymbol{r}',\omega)\delta V(\boldsymbol{r}')$$

$$\Pi_0(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{i}{\hbar}\lim_{\eta\to 0}\int_0^\infty d\tau\; \exp\left(i(\omega+i\eta)\tau\right)\langle[\hat{\rho}_I(\boldsymbol{r},\tau),\hat{\rho}(\boldsymbol{r}')]\rangle_0$$

- response in the particle current density $\delta j_z(\boldsymbol{r},t) = \delta j_z(\boldsymbol{r})\exp(-i\omega t)$

$$\delta j_z(\boldsymbol{r}) = \int d^3r'\, \tilde{\Pi}_0(\boldsymbol{r},\boldsymbol{r}',\omega)\delta V(\boldsymbol{r}')$$

$$\tilde{\Pi}_0(\boldsymbol{r},\boldsymbol{r}',\omega) = \frac{i}{\hbar}\lim_{\eta\to 0}\int_0^\infty d\tau\; \exp\left(i(\omega+i\eta)\tau\right)\langle[\hat{j}_{zI}(\boldsymbol{r},\tau),\hat{\rho}(\boldsymbol{r}')]\rangle_0$$

- charge-charge correlation function

$$\Pi_0(\mathbf{r},\mathbf{r}',\omega) = 2\lim_{\eta\to 0}\sum_{\alpha,\alpha'}\frac{\bar{n}_\alpha-\bar{n}_{\alpha'}}{E_\alpha-E_{\alpha'}+\hbar(\omega+i\eta)}\varphi^*_\alpha(\mathbf{r})\varphi_{\alpha'}(\mathbf{r})\varphi^*_{\alpha'}(\mathbf{r}')\varphi_\alpha(\mathbf{r}')$$

- current-charge correlation function

$$\tilde{\Pi}_0(\mathbf{r},\mathbf{r}',\omega) = -2\lim_{\eta\to 0}\sum_{\alpha,\alpha'}\frac{\bar{n}_\alpha-\bar{n}_{\alpha'}}{E_\alpha-E_{\alpha'}+\hbar(\omega+i\eta)} \times\frac{\hbar}{2im}\left(\varphi^*_\alpha(\mathbf{r})\frac{\partial}{\partial z}\varphi_{\alpha'}(\mathbf{r})-\varphi_{\alpha'}(\mathbf{r})\frac{\partial}{\partial z}\varphi^*_\alpha(\mathbf{r})\right)\varphi^*_{\alpha'}(\mathbf{r}')\varphi_\alpha(\mathbf{r}')$$

Response functions can be computed using the one-particle scattering states of the quantum system under external static bias.

Assumptions for open quantum systems

- consider perturbation in effective potential
- retardation effects can be neglected ($\omega \ll c/2d \approx 10^{16} Hz$)
- planar structure: $\delta V(\boldsymbol{r}, t) = \delta V(z, t)$
- no phase coherence between contacts and scattering area $\Pi_0(|z| < d, |z'| > d) = 0$

⇒ only z dependence remains on finite domain

- changes in particle density

$$\delta\rho(|z| \leq d) = \int_{-d}^{d} dz' \Pi_0(z, z', \omega)\delta V(z')$$

- changes in particle current density

$$\delta j_z(|z| \leq d) = \int_{-d}^{d} dz' \tilde{\Pi}_0(z, z', \omega)\delta V(z')$$

- continuity equation

$$\frac{\partial}{\partial z}\delta j_z(z, t) + \frac{\partial}{\partial t}\delta\rho(z, t) = 0$$

$$\Delta \delta V(z) = -\frac{e^2}{\kappa_s} \delta\rho(z)$$

with b.c. (good screening in contacts) $\delta V(z \leq -d) = 0$
$\delta V(z \geq d) = -e\delta U$

- integral equation

$$\delta V(z) = \delta V_0(z) + \int_{-d}^{d} dz' \int_{-d}^{d} dz'' v_0(z,z')\Pi_0(z',z'',\omega)\delta V(z'')$$

  with $v_0(z,z')$ the Green's function and $\delta V_0(z)$ a special solution.
- discretized form $\Rightarrow$ matriceal equation

$$\delta \boldsymbol{V} = (\mathbf{1} - \boldsymbol{v}_0 \boldsymbol{\Pi}_0)^{-1} \delta \boldsymbol{V}_0$$

One can compute microscopically the dynamic changes in the effective potential.

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with $v_0(z,z')$ the Green's function and $\delta V_0(z)$ a special solution.

- discretized form $\Rightarrow$ matriceal equation

$$\delta \boldsymbol{V} = (\mathbf{1} - \boldsymbol{v}_0\boldsymbol{\Pi}_0)^{-1}\,\delta\boldsymbol{V}_0$$

One can compute microscopically the dynamic changes in the effective potential, and also in the particle density and in the particle current density.

$$\delta\boldsymbol{\rho} = \boldsymbol{\Pi}_0\delta\boldsymbol{V} = \boldsymbol{\Pi}_0\,(\mathbf{1} - \boldsymbol{v}_0\boldsymbol{\Pi}_0)^{-1}\,\delta\boldsymbol{V}_0$$

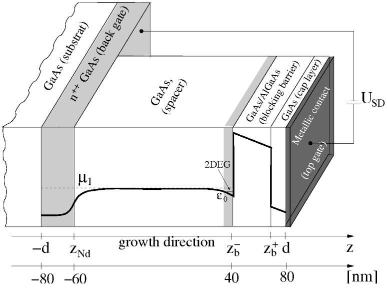
$$\delta\boldsymbol{j}_z = \tilde{\boldsymbol{\Pi}}_0\delta\boldsymbol{V} = \tilde{\boldsymbol{\Pi}}_0\,(\mathbf{1} - \boldsymbol{v}_0\boldsymbol{\Pi}_0)^{-1}\,\delta\boldsymbol{V}_0$$

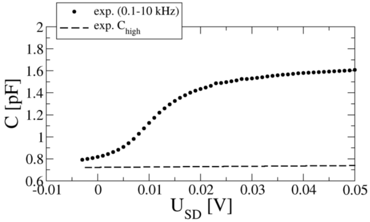
$$Y(\omega) = \frac{1}{Z(\omega)} = \frac{\delta I(t)}{\delta U(t)}$$

with

$$\begin{aligned} \delta I(t) &= -eA\delta j_z(-d) = -eA\delta j_z(d) \\ \delta U(t) &= \delta U e^{-i(\omega+i\eta)t} \end{aligned}$$

V.T. Dolgopolov, W Hansen, et al. Phys. Low-Dim. Struct. **6**, 1, (1996)





- $\psi^{(1)}(\epsilon, z \geq d) = 0, \quad \psi^{(2)}(\epsilon, z \leq -d) = 0.$
- $\psi^{(1)}(\epsilon, z)\psi^{(2)}(\epsilon', z) \sim 0$

$\Rightarrow \delta j_z = \delta j_z^{(1)} + \delta j_z^{(2)}$

- continuity equation for each component
  $\Rightarrow \delta j_z^{(1)}(-d) = -i\omega \int_{-d}^{d} dz \int_{-d}^{d} dz' \Pi_0^{(11)}(z, z', \omega)\delta V(z')$
  $\Rightarrow$ only charge-charge correlation function is needed

- quantum system in contact with one reservoir only: $s = 1$

Small frequency expansion: first $\omega \ll 1$ after that $\eta \to 0$,

$$\Pi_0^{(ss)}(z,z',\omega) = P_0^{(s)}(z,z') + i\omega P_1^{(s)}(z,z')$$

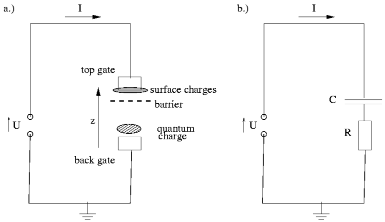
with $P_0^{(s)}(z,z'), P_1^{(s)}(z,z') \in R$

$$Y(\omega \to 0) \approx -i\omega(Y_1 + i\omega Y_2)$$

with $Y_1,\ Y_2 \in R$

$$U(t) = U_G + \delta U e^{-i\omega t}$$

$$\Rightarrow Z_{sg}(\omega) = R + i\frac{1}{\omega C}$$



admittance of RC circuit

$$Y_{sg}(\omega) = \frac{1}{Z_{sg}(\omega)} = \frac{RC^2\omega^2 - i\omega C}{1 + R^2C^2\omega^2}$$

small frequency

$$Y(\omega \to 0) = RC^2\omega^2 - i\omega C$$
$$Y(\omega \to 0) \approx Y_2\omega^2 - i\omega Y_1$$

$\Rightarrow Y_1 = C$ and $Y_2 = RC^2$

quantum mechanical expresions for the equivalent circuit elements

U. Wulf, P.N. Racec and E. R. Racec, Phys. Rev. **B 75**, 075320, (2007) normalized form: $\bar{Y} = YY_2/Y_1^2$

$\bar{\omega} = \omega/\omega_0$

with normalization factors:

$$Y_1 = -\lim_{\omega\to 0} Im[Y(\omega)]/\omega$$

$$Y_2 = \lim_{\omega\to 0} Re[Y(\omega)]/\omega^2$$

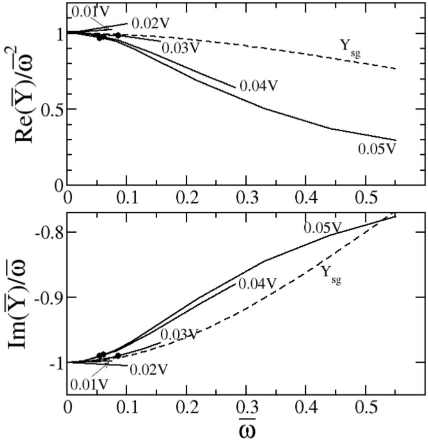
$$\omega_0 = Y_1/Y_2$$

admittance of RC circuit

$$\bar{Y}_{sg}(\bar{\omega}) = -i\bar{\omega}\left(\frac{1}{1+\bar{\omega}^2} + i\frac{\bar{\omega}}{1+\bar{\omega}^2}\right)$$

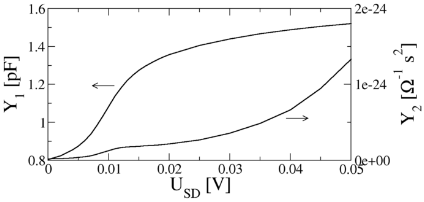
low frequency

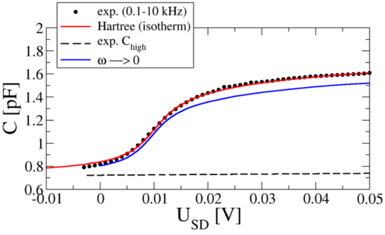
$$\bar{Y}(\bar{\omega}\to 0) = -i\bar{\omega}(1+i\bar{\omega})$$



U. Wulf, P.N. Racec and E. R. Racec, Phys. Rev. **B 75**, 075320, (2007)

systematic deviations from RC-circuit with frequency independent R and C





U. Wulf, P.N. Racec and E. R. Racec, Phys. Rev. **B 75**, 075320, (2007)

Physical interpretation:

- classical:

$$Y(\omega) = -i\omega C_e + \omega^2 C_e^2 R + O(\omega^3)$$

  with C_e the electrostatic and geometrical capacitance

- nanostructure:

$$Y(\omega) = -i\omega C_\mu + \omega^2 C_\mu^2 R_q + O(\omega^3)$$

  with C_μ the electrochemical capacitance
  R_q the charge relaxation resistance

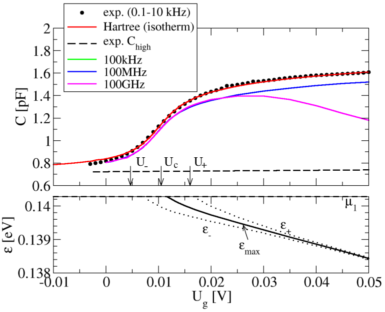
M. Büttiker et al., Phys. Lett. A. 180, 364 (1993)

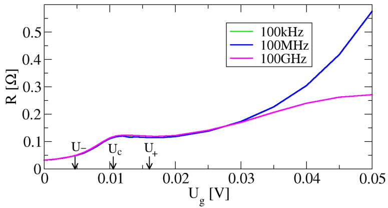
- quantum mechanical model for the admittance of effectively 1D open systems
  - linear response theory and random-phase approximation
  - the model is valid also for large static bias and for high frequencies
- the dynamic changes in the effective potential are calculated microscopically from the charge-charge correlation function
- as an example, a quantum capacitor is described
  - a small-signal equivalent circuit with frequency-independent elements is not suitable for high frequencies

- Green's function $v_0(z,z') = -(e^2/2\kappa_s)[|z-z'| + zz'/d - d]$
- density-density correlation function

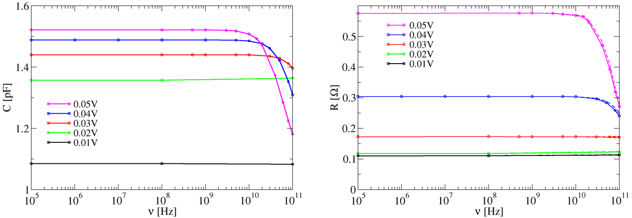
$$\Pi_0(z,z',\omega) = \sum_{s,s'=1}^{2} \lim_{\eta\to 0} \int_{V_s}^{\infty} d\epsilon \int_{V_{s'}}^{\infty} d\epsilon' \, \frac{F^{(ss')}(z,z',\epsilon,\epsilon')}{\epsilon - \epsilon' + \hbar(\omega + i\eta)}$$

$$\begin{aligned} F^{(ss')}(z,z',\epsilon,\epsilon') &= 2\frac{m^*}{2\pi\beta\hbar^2} g_s(\epsilon) g_{s'}(\epsilon') \ln\left\{ \frac{1+\exp\left(\beta(\mu_s-\epsilon)\right)}{1+\exp\left(\beta(\mu_{s'}-\epsilon')\right)} \right\} \\ &\times \left(\psi^{(s)}(\epsilon,z)\right)^* \psi^{(s')}(\epsilon',z) \left(\psi^{(s')}(\epsilon',z')\right)^* \psi^{(s)}(\epsilon,z'). \end{aligned}$$

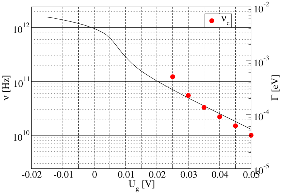




- bias dependent capacitance and resistance
- frequency dependence for higher frequencies



- width of resonance $\Gamma = \hbar 2\pi \nu_c$: $\nu > \nu_c \Rightarrow \tau < \tau_c$



P.N. Racec and U. Wulf, Mat. Sci. Eng. C 26, 876 (2006)