The microscopic origin of the macroscopic dielectric permittivity of crystals

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Microscopic origin of macroscopic dielectric properties (1)

In a dielectric material, the presence of an electric field causes the nuclear and electronic charges to slightly separate, inducing a local electric dipole.

This generates an induced response inside the material (reorganization of the electronic density), *screening* the applied field.
**Microscopic origin of macroscopic dielectric properties (2)**

- **Dielectric material:** can polarize in presence of external fields

<table>
<thead>
<tr>
<th>density</th>
<th>electric field</th>
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<tbody>
<tr>
<td>$\nu$</td>
<td>$D, \ \text{div} \ D = 4\pi \nu$</td>
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<tr>
<td>$\delta \rho$</td>
<td>$P, \ \text{div} \ P = 4\pi \delta \rho$</td>
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<tr>
<td>$\rho$</td>
<td>$E, \ \text{div} \ E = 4\pi \rho$</td>
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- **Constitutive equation:** $\varepsilon_M = 3 \times 3$ symmetric real matrix with $\varepsilon_M \geq 1$

\[
D = \varepsilon_M E \iff P = (\varepsilon_M - 1)E = (1 - \varepsilon_M^{-1})D
\]

- **Time-dependent fields:** the response of the material is not instantaneous, but given by a convolution with some response function. With $E(t) = -\nabla W(t)$ where $W(t)$ is the macroscopic potential,

\[-\text{div} \left( \varepsilon_M(\omega) \nabla \widehat{W}(\omega) \right) = 4\pi \widehat{\nu}(\omega)\]
Another motivation: Opto-electronical properties

- **Photovoltaic effect**: convert light into electric current/voltage
  - Mechanism: promotion of valence electrons into excited states
  - Compute band gaps in photovoltaic materials
  - Need for methods going beyond standard ground-state approaches

- Reference numerical approach: **GW method** → horrible equations...

  - Dyson equation \( G(12) = G^{(0)}(12) + \int d(34)G^{(0)}(13)\Sigma(34)G(42) \)
  - Self-energy \( \Sigma(12) = i \int d(34)W(1^+3)G(14)\Gamma(42; 3) \)

  **Screened interaction**

  \[
  W(12) = \nu(12) + \int d(34)W(13)P(34)\nu(42)
  \]

  - Irreducible polarization \( P(12) = -i \int d(34)G(23)G(42)\Gamma(34; 1) \)
  - Vertex function \( \Gamma(12; 3) = \delta(12)\delta(13) + \int d(4567) \frac{\delta \Sigma(12)}{\delta G(45)} G(46)G(75)\Gamma(67; 3) \)
Outline

Some background material
- Description of perfect crystals
- Crystals with defects: static picture

Time evolution of defects in crystals
- Response to an effective potential
- Well-posedness of the nonlinear Hartree dynamics
- Frequency dependent macroscopic dielectric permittivity

Elements of electronic structure theory
Some elements on trace-class operators

- Compact self-adjoint operator $A = \sum_{i=1}^{+\infty} \lambda_i \langle \phi_i | \phi_i \rangle$ with $\lambda_i \to 0$

- The operator $A$ is called trace-class ($A \in \mathcal{S}_1$) if $\sum_{i=1}^{+\infty} |\lambda_i| < \infty$. Its density

$$\rho_A(x) = \sum_{i=1}^{+\infty} \lambda_i |\phi_i(x)|^2$$

belongs to $L^1(\mathbb{R}^3)$ and

$$\text{Tr}(A) := \sum_{i=1}^{+\infty} \lambda_i = \sum_{i=1}^{+\infty} \langle e_i | A | e_i \rangle = \int_{\mathbb{R}^3} \rho_A$$

- $A$ is Hilbert-Schmidt ($A \in \mathcal{S}_2$) if $A^*A \in \mathcal{S}_1$, i.e. $\sum_{i \geq 1} |\lambda_i|^2 < \infty$. If $A$ is self-adjoint, its integral kernel is in $L^2(\mathbb{R}^3 \times \mathbb{R}^3)$

$$A(x, y) = \sum_{i \geq 1} \lambda_i \overline{\phi_i(x)} \phi_i(y).$$
Density operators for a finite system of $N$ electrons in $\mathbb{R}^3$

- Bounded, self-adjoint operator on $L^2(\mathbb{R}^3)$ such that $0 \leq \gamma \leq 1$ and $\text{Tr}(\gamma) = N$. In some orthonormal basis of $L^2(\mathbb{R}^3)$,

$$
\gamma = \sum_{i=1}^{+\infty} n_i |\phi_i\rangle \langle \phi_i|, \quad 0 \leq n_i \leq 1, \quad \sum_{i=1}^{+\infty} n_i = N
$$

- For the Slater determinant $\psi(x_1, \ldots, x_N) = (N!)^{-1/2} \det(\phi_i(x_j))_{1 \leq i,j \leq N}$,

$$
\gamma_\psi = \sum_{i=1}^{N} |\phi_i\rangle \langle \phi_i|
$$

- Electronic density $\rho_\gamma(x) = \sum_{i=1}^{+\infty} n_i |\phi_i(x)|^2$ with $\rho_\gamma \geq 0$ and $\int_{\mathbb{R}^3} \rho_\gamma = N$.

- Kinetic energy $T(\gamma) = \frac{1}{2} \text{Tr}(|\nabla|\gamma|\nabla|) = \frac{1}{2} \sum_{i=1}^{+\infty} n_i \|\nabla \phi_i\|^2_{L^2(\mathbb{R}^3)}$
The Hartree model for finite systems

- **Hartree energy** $E_{\rho_{\text{nuc}}}^{\text{Hartree}}(\gamma) = \text{Tr} \left( -\frac{1}{2} \Delta \gamma \right) + \frac{1}{2} D(\rho_\gamma - \rho_{\text{nuc}}, \rho_\gamma - \rho_{\text{nuc}})$

where

$$D(f, g) = \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{f(x) g(x')}{|x - x'|} \, dx \, dx' = 4\pi \int_{\mathbb{R}^3} \frac{\hat{f}(k) \hat{g}(k)}{|k|^2} \, dk$$

is the classical Coulomb interaction, defined for $f, g \in L^{6/5}(\mathbb{R}^3)$, but which can be extended to

$$\mathcal{C} = \left\{ f \in \mathscr{S}'(\mathbb{R}^3) \mid \hat{f} \in L^1_{\text{loc}}(\mathbb{R}^3), \ | \cdot |^{-1} \hat{f}(\cdot) \in L^2(\mathbb{R}^3) \right\}$$

**Variational formulation**

$$\inf \left\{ E_{\rho_{\text{nuc}}}^{\text{Hartree}}(\gamma), \, \gamma \in \mathcal{S}(L^2(\mathbb{R}^3)), \, 0 \leq \gamma \leq 1, \, \text{Tr}(\gamma) = N, \, \text{Tr}(-\Delta \gamma) < \infty \right\}$$

- More general models of density functional theory: correction term $E_{\text{xc}}(\gamma)$

Euler-Lagrange equations for the Hartree model

Nonlinear eigenvalue problem, $\varepsilon_F$ Lagrange multiplier of $\text{Tr}(\gamma) = N$

$$
\begin{align*}
\gamma^0 &= \sum_{i=1}^{+\infty} n_i |\phi_i\rangle\langle\phi_i|, \\
\rho^0(x) &= \sum_{i=1}^{+\infty} n_i |\phi_i(x)|^2, \\
H^0 \phi_i &= \varepsilon_i \phi_i, \\
\langle\phi_i, \phi_j\rangle &= \delta_{ij}, \\
n_i &= \begin{cases} 
1 & \text{if } \varepsilon_i < \varepsilon_F \\
\in [0, 1] & \text{if } \varepsilon_i = \varepsilon_F \\
0 & \text{if } \varepsilon_i > \varepsilon_F 
\end{cases}, \\
\sum_{i=1}^{+\infty} n_i &= N, \\
H^0 &= -\frac{1}{2} \Delta + V^0, \\
-\Delta V^0 &= 4\pi (\rho^{\text{nuc}} - \rho^0).
\end{align*}
$$

When $\varepsilon_N < \varepsilon_{N+1}$ (gap):

$$
\begin{align*}
\gamma^0 &= 1_{(-\infty, \varepsilon_F]}(H^0), \\
H^0 &= -\frac{1}{2} \Delta + V^0, \\
-\Delta V^0 &= 4\pi (\rho^{\text{nuc}} - \rho^0),
\end{align*}
$$
The Hartree model for crystals (1)

- **Thermodynamic limit**, periodic nuclear density $\rho_{\text{nuc}}$, lattice $R \simeq (a\mathbb{Z})^3$ with unit cell $\Gamma$, reciprocal lattice $R^* \simeq \left( \frac{2\pi}{a} \mathbb{Z} \right)^3$ with unit cell $\Gamma^*$

- **Bloch-Floquet transform**: unitary $L^2(\mathbb{R}^3) \to \bigoplus_{\Gamma^*} L^2_{\text{per}}(\Gamma) dq$

  $$f_q(x) = \sum_{R \in \mathcal{R}} f(x + R) e^{-i q \cdot (x+R)} = \frac{(2\pi)^{3/2}}{|\Gamma|} \sum_{K \in R^*} \hat{f}(q + K) e^{iK \cdot x}$$

- Any operator commuting with the spatial translations $\tau_R (R \in \mathcal{R})$ can be decomposed as $(Af)_q = A_q f_q$, and $\sigma(A) = \bigcup_{q \in \Gamma^*} \sigma(A_q)$

- **Bloch matrices**: $A_{K,K'}(q) = \langle e_K, A_q e_{K'} \rangle_{L^2_{\text{per}}(\Gamma)}$, $e_K(x) = |\Gamma|^{-1/2} e^{iK \cdot x}$

  $$\mathcal{F}(Av)(q + K) = \sum_{K' \in \mathcal{R}^*} A_{K,K'}(q) \mathcal{F}v(q + K')$$


The Hartree model for crystals (2)

Nonlinear eigenvalue problem

\[
\begin{align*}
\gamma^0_{\text{per}} &= 1_{(-\infty, \varepsilon_F]}(H^0_{\text{per}}), & \rho^0_{\text{per}} &= \rho_{\gamma^0_{\text{per}}}, \\
H^0_{\text{per}} &= -\frac{1}{2}\Delta + V^0_{\text{per}}, \\
-\Delta V^0_{\text{per}} &= 4\pi(\rho^\text{nuc}_{\text{per}} - \rho^0_{\text{per}}), & \int_{\Gamma} \rho^0_{\text{per}} = \int_{\Gamma} \rho^\text{nuc}_{\text{per}} = N
\end{align*}
\]

More explicit expressions using the Bloch decomposition

\[
(H^0_{\text{per}})_q = -\frac{1}{2}\Delta - iq \cdot \nabla + \frac{|q|^2}{2} + V^0_{\text{per}} = \sum_{n=1}^{+\infty} \varepsilon_{n,q} |u_{n,q}\rangle \langle u_{n,q}|
\]

\[
(\gamma^0_{\text{per}})_q = \sum_{n=1}^{+\infty} 1_{\{\varepsilon_{n,q} \leq \varepsilon_F\}} |u_{n,q}\rangle \langle u_{n,q}|
\]

Fermi level obtained from $N = \frac{1}{|\Gamma^*|} \sum_{n=1}^{+\infty} |\{q \in \Gamma^* | \varepsilon_{n,q} \leq \varepsilon_F\}|$
The Hartree model for crystals (3)

The spectrum of the periodic Hamiltonian is composed of bands

$$\sigma(H) = \bigcup_{n \geq 1} \left[ \Sigma_n^-, \Sigma_n^+ \right], \quad \Sigma_n^- = \min_{q \in \Gamma^*} \varepsilon_{n,q}, \quad \Sigma_n^+ = \max_{q \in \Gamma^*} \varepsilon_{n,q}$$

Assume in the sequel that

$$g = \Sigma_{N+1}^- - \Sigma_N^+ > 0 \quad \text{(insulator)}$$

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Grenoble, April 2013 13 / 31**
Defects in crystals
Local defects (1)

- Local perturbation: nuclear charge defect $\rho_{\text{per}}^{\text{nuc}} + \nu$

- Expected ground state $\gamma = \gamma_{\text{per}}^{0} + Q\nu$ with $Q\nu$ ‘local’
Local defects (2)

- Local perturbation: nuclear charge defect $\rho_{\text{nuc}} + \nu$

- Expected ground state $\gamma = \gamma_{\text{per}}^0 + Q_{\nu}$ with $Q_{\nu}$ “local”
Defects in crystals (1)

- A thermodynamic limit shows that $Q_\nu$ can be thought of as some defect state embedded in the periodic medium

$$Q_\nu = \operatorname{argmin}_{Q \in Q} \left\{ \operatorname{Tr}_0 (H^0_{\text{per}} Q) - \int_{\mathbb{R}^3} \rho_Q (\nu \star | \cdot |^{-1}) + \frac{1}{2} D(\rho_Q, \rho_Q) \right\}$$

where, defining $Q^{--} = \gamma^0_{\text{per}} Q \gamma^0_{\text{per}}$ and $Q^{++} = (1 - \gamma^0_{\text{per}}) Q (1 - \gamma^0_{\text{per}})$,

$$Q = \left\{ Q^* = Q, \ (1 - \Delta)^{1/2} Q \in \mathbb{S}_2, \ (1 - \Delta)^{1/2} Q^{\pm\pm} (1 - \Delta)^{1/2} \in \mathbb{S}_1 \right\}$$

- Generalized trace $\operatorname{Tr}_0(Q) = \operatorname{Tr}(Q^{++}) + \operatorname{Tr}(Q^{--})$

- Density $\rho_Q \in L^2(\mathbb{R}^3) \cap \mathcal{C}$

Defects in crystals (2)

Definition of the embedding energy

\[
\text{Tr}_0((H^0_{\text{per}} - \varepsilon_F)Q) := \text{Tr}(\|H^0_{\text{per}} - \varepsilon_F\|^{1/2}(Q^{++} - Q^{--})\|H^0_{\text{per}} - \varepsilon_F\|^{1/2})
\]

[CL, Theorem 1]

Let \( \nu \) such that \( (\nu \star | \cdot |^{-1}) \in L^2(\mathbb{R}^3) + C' \). Then, there exists at least one minimizer \( Q_{\nu,\varepsilon_F} \), and all the minimizers share the same density \( \rho_{\nu,\varepsilon_F} \). In addition, \( Q_{\nu,\varepsilon_F} \) is solution to the self-consistent equation

\[
Q_{\nu,\varepsilon_F} = 1_{(-\infty,\varepsilon_F)}(H^0_{\text{per}} + (\rho_{\nu,\varepsilon_F} - \nu) \star | \cdot |^{-1}) - 1_{(-\infty,\varepsilon_F]}(H^0_{\text{per}}) + \delta,
\]

where \( \delta \) is a finite-rank self-adjoint operator on \( L^2(\mathbb{R}^3) \) such that \( 0 \leq \delta \leq 1 \) and \( \text{Ran}(\delta) \subset \text{Ker}(H^0_{\text{per}} + (\rho_{\nu,\varepsilon_F} - \nu) \star | \cdot |^{-1} - \varepsilon_F) \).

When \( \nu \) is sufficiently small, \( \delta = 0 \) and the minimizer is unique.
Time evolution of defects in crystals: effective perturbations
The time-dependent Hartree dynamics

- **Finite system** described by the density matrix $\gamma(t)$, von Neumann equation

$$i \frac{d\gamma(t)}{dt} = \left[H^0_{\gamma(t)}, \gamma(t)\right], \quad H^0_{\gamma} = -\frac{1}{2} \Delta + V_{\text{nuc}} + v_c(\rho_{\gamma})$$

- When a perturbation $v(t)$ is added, the dynamics is modified as

$$i \frac{d\gamma(t)}{dt} = \left[H^0_{\gamma(t)} + v(t), \gamma(t)\right],$$

- **Formal thermodynamic limit**: state $\gamma(t) = \gamma^0_{\text{per}} + Q(t)$ and dynamics

$$i \frac{d\gamma}{dt} = \left[H^\nu_{\gamma}, \gamma\right], \quad H^\nu_{\gamma}(t) = H^0_{\text{per}} + v_c(\rho_{Q(t)} - \nu(t))$$


Defects in a time-dependent setting: the dynamics

Classical formulation: nonlinear dynamics

\[ i \frac{dQ(t)}{dt} = \left[ H^0_{\text{per}} + v_c(\rho Q(t) - \nu(t)), \gamma^0_{\text{per}} + Q(t) \right] \]

Denote \( U_0(t) = e^{-itH^0_{\text{per}}} \) the free evolution.

Mild formulation for an effective potential \( \nu(t) \)

\[ Q(t) = U_0(t)Q^0U_0(t)^* - i \int_0^t U_0(t - s)[\nu(s), \gamma^0_{\text{per}} + Q(s)]U_0(t - s)^* \, ds \]

Mild formulation for the nonlinear dynamics

Replace \( \nu(s) \) by \( \nu_c(\rho Q(s) - \nu(s)) \) in the above formula
Well-posedness of the mild formulation

If initially $Q(0) \in Q$, the Banach space allowing to describe local defects in crystals, does $Q(t) \in Q$?

[CS12, Proposition 1]

The integral equation has a unique solution in $C^0(\mathbb{R}_+, Q)$ for $Q^0 \in Q$ and $v = v_c(\rho)$ with $\rho \in L^1_{\text{loc}}(\mathbb{R}_+, L^2(\mathbb{R}^3) \cap C)$. In addition, $\text{Tr}_0(Q(t)) = \text{Tr}_0(Q^0)$, and, if $-\gamma^0_{\text{per}} \leq Q^0 \leq 1 - \gamma^0_{\text{per}}$, then $-\gamma^0_{\text{per}} \leq Q(t) \leq 1 - \gamma^0_{\text{per}}$.

This result is based on a series of technical results:

- boundedness of the potential: $v \in L^1_{\text{loc}}(\mathbb{R}_+, L^\infty(\mathbb{R}^3))$
- stability of time evolution: \[ \frac{1}{\beta} \| Q \|_Q \leq \| U_0(t)QU_0(t)^* \|_Q \leq \beta \| Q \|_Q \]
- commutator estimates with $\gamma^0_{\text{per}}$: $\| i[v, \gamma^0_{\text{per}}] \|_Q \leq C_{\text{com}} \| v \|_{C'}$
- commutator estimates in $Q$: $\| i[v_c(\varrho), Q] \|_Q \leq C_{\text{com}, Q} \| \varrho \|_{L^2 \cap C} \| Q \|_Q$
Dyson expansion and linear response

Response at all orders (formally): $Q(t) = U_0(t)Q^0U_0(t)^* + \sum_{n=1}^{+\infty} Q_{n,v}(t)$

$Q_{1,v}(t) = -i \int_0^t U_0(t-s) [v(s), \gamma_{\text{per}}^0 + U_0(s)Q^0U_0(s)^*] U_0(t-s)^* \, ds,$

$Q_{n,v}(t) = -i \int_0^t U_0(t-s) [v(s), Q_{n-1,v}(s)] U_0(t-s)^* \, ds \quad \text{for } n \geq 2$

Obtained by plugging the formal decomposition into the integral equation

[CS12, Proposition 5]

Under the previous assumptions, $Q_{n,v} \in C^0(\mathbb{R}_+, Q)$ with $\text{Tr}_0(Q_{n,v}(t)) = 0$,

$$\|Q_{n,v}(t)\|_Q \leq \beta \frac{1 + \|Q^0\|_Q}{n!} \left( C \int_0^t \|\rho(s)\|_{L^2 \cap C} \, ds \right)^n.$$

The formal expansion therefore converges in $Q$, uniformly on any compact subset of $\mathbb{R}_+$, to the unique solution in $C^0(\mathbb{R}_+, Q)$ of the integral equation.
Definition of the polarization (1)

- **Aim**: Justify the Adler-Wiser formula for the polarization matrix

- **Damped linear response**: standard linear response as $\eta \to 0$

\[
Q^n_{1,\nu}(t) = -i \int_{-\infty}^{t} U_0(t-s) \left[ \nu(s), \gamma^0_{\text{per}} \right] U_0(t-s)^* e^{-\eta(t-s)} ds
\]

- polarization operator $\chi^\eta_0 : \begin{cases} L^1(\mathbb{R}, C') &\rightarrow& C^0_b(\mathbb{R}, L^2(\mathbb{R}^3) \cap C) \\ \nu &\mapsto& \rho Q^n_{1,\nu} \end{cases}$

- linear response operator $\mathcal{E}^\eta = \nu_c^{1/2} \chi_0 \nu_c^{1/2}$ acting on $L^1(\mathbb{R}, L^2(\mathbb{R}^3))$

\[
\langle f_2, \mathcal{E}^\eta f_1 \rangle_{L^2(L^2)} = \int_{\mathbb{R}} \langle \mathcal{F}_t f_2(\omega), \mathcal{E}^\eta(\omega) \mathcal{F}_t f_1(\omega) \rangle_{L^2(\mathbb{R}^3)} d\omega
\]

- Bloch decomposition: for a.e. $(\omega, q) \in \mathbb{R} \times \Gamma^*$ and any $K \in \mathcal{R}^*$,

\[
\mathcal{F}_{t,x} (\mathcal{E}^\eta f) (\omega, q + K) = \sum_{K' \in \mathcal{R}^*} \mathcal{E}^\eta_{K,K'}(\omega, q) \mathcal{F}_{t,x} f(\omega, q + K')
\]


Definition of the polarization (2)

[CS12, Proposition 7]

The Bloch matrices of the damped linear response operator $\mathcal{E}^\eta$ read

$$\mathcal{E}^\eta_{K,K'}(\omega, q) = \frac{1}{\Gamma} \frac{|q + K'|}{|q + K|} T^\eta_{K,K'}(\omega, q),$$

where the continuous functions $T^\eta_{K,K'}$ are uniformly bounded:

$$T^\eta_{K,K'}(\omega, q) = \sum \int_{\Gamma^*} \frac{\langle u_m, q', e^{-iK' \cdot x} u_n, q+q' \rangle_{L^2_{\text{per}}} \langle u_n, q+q', e^{iK' \cdot x} u_m, q' \rangle_{L^2_{\text{per}}} dq'}{\varepsilon_{n,q+q'} - \varepsilon_{m,q'} - \omega - i\eta}$$

(the sum is over $1 \leq n \leq N < m$ and $1 \leq m \leq N < n$)

- The Bloch matrices of the standard linear response are recovered as $\eta \to 0$, the convergence being in $\mathcal{S}'(\mathbb{R} \times \mathbb{R}^3)$

- Static polarizability recovered in some adiabatic limit
Time evolution of defects in crystals: nonlinear dynamics
Time-dependent Hartree dynamics for defects

Well-posedness of the mild formulation

For $\nu \in L^1_{\text{loc}}(\mathbb{R}^+, L^2(\mathbb{R}^3)) \cap W^{1,1}_{\text{loc}}(\mathbb{R}^+, C)$, and $-\gamma_0^\text{per} \leq Q^0 \leq 1 - \gamma_0^\text{per}$ with $Q^0 \in Q$, the dynamics

$$Q(t) = U_0(t)Q^0U_0(t)^* - i \int_0^t U_0(t-s) \left[ v_c(\rho_{Q(s)} - \nu(s)), \gamma^0_\text{per} + Q(s) \right] U_0(t-s)^* ds$$

has a unique solution in $C^0(\mathbb{R}^+, Q)$. For all $t \geq 0$, $\text{Tr}_0(Q(t)) = \text{Tr}_0(Q^0)$ and $-\gamma_0^\text{per} \leq Q(t) \leq 1 - \gamma_0^\text{per}$.

- Idea of the proof: (i) short time existence and uniqueness by a fixed-point argument; (ii) extension to all times by controlling the energy

$$\mathcal{E}(t, Q) = \text{Tr}_0(H^0_{\text{per}} Q) - D(\rho_Q, \nu(t)) + \frac{1}{2} D(\rho_Q, \rho_Q)$$

- Classical solution well posed under stronger assumptions on $Q^0, \nu$
Macroscopic dielectric permittivity (1)

Starting from $Q^0 = 0$, the nonlinear dynamics can be rewritten as

$$Q(t) = Q_{1,v_c(\rho_Q - \nu)}(t) + \tilde{Q}_{2,v_c(\rho_Q - \nu)}(t)$$

In terms of electronic densities: $[(1 + L)(\nu - \rho_Q)](t) = \nu(t) - r_2(t)$

Properties of the operator $L$

For any $0 < \Omega < g$, the operator $L$ is a non-negative, bounded, self-adjoint operator on the Hilbert space

$$\mathcal{H}_\Omega = \left\{ \varrho \in L^2(\mathbb{R}, \mathbb{C}) \mid \text{supp}(F_{t,x}\varrho) \subset [-\Omega, \Omega] \times \mathbb{R}^3 \right\},$$

endowed with the scalar product

$$\langle \varrho_2, \varrho_1 \rangle_{L^2(\mathbb{C})} = 4\pi \int_{-\Omega}^{\Omega} \int_{\mathbb{R}^3} \frac{\overline{F_{t,x}\varrho_2(\omega, k)} F_{t,x}\varrho_1(\omega, k)}{|k|^2} \, d\omega \, dk.$$ 

Hence, $1 + L$, considered as an operator on $\mathcal{H}_\Omega$, is invertible.
Macroscopic dielectric permittivity (2)

- **Linearization:** given \( \nu \in \mathcal{H}_\Omega \), find \( \rho_\nu \) such that \((1 + \mathcal{L})(\nu - \rho_\nu) = \nu\)

- **Homogenization limit:** spread the charge as \( \nu_\eta(t, x) = \eta^3 \nu(t, \eta x) \) and consider the rescaled potential
  \[
  W^\eta_\nu(t, x) = \eta^{-1} v_c(\nu_\eta - \rho_{\nu_\eta})(t, \eta^{-1} x)
  \]

When \( \mathcal{L} = 0 \), the potential is \( W^\eta_\nu = v_c(\nu) \)

[CS12, Proposition 14]
The rescaled potential \( W^\eta_\nu \) converges weakly in \( \mathcal{H}_\Omega \) to the unique solution \( W_\nu \) in \( \mathcal{H}_\Omega \) to the equation

\[
- \text{div} \left( \varepsilon_M(\omega) \nabla [\mathcal{F}_t W_\nu](\omega, \cdot) \right) = 4\pi [\mathcal{F}_t \nu](\omega, \cdot)
\]

where \( \varepsilon_M(\omega) \) (for \( \omega \in (-g, g) \)) is a smooth mapping with values in the space of symmetric \( 3 \times 3 \) matrices, and satisfying \( \varepsilon_M(\omega) \geq 1 \).

- The matrix \( \varepsilon_M(\omega) \) can be expressed using the Bloch decomposition
Perspectives
Perspectives and open issues

• **Metallic** systems (no gap: many estimates break down)

• **Longtime** behavior of the defect

• Influence of **electric and magnetic fields** (rather than a local perturbation as was the case here)

• Interaction of electronic defects with **phonons** (lattice vibrations)

• **GW methods** (the polarization matrix enters the definition of the self-energy)