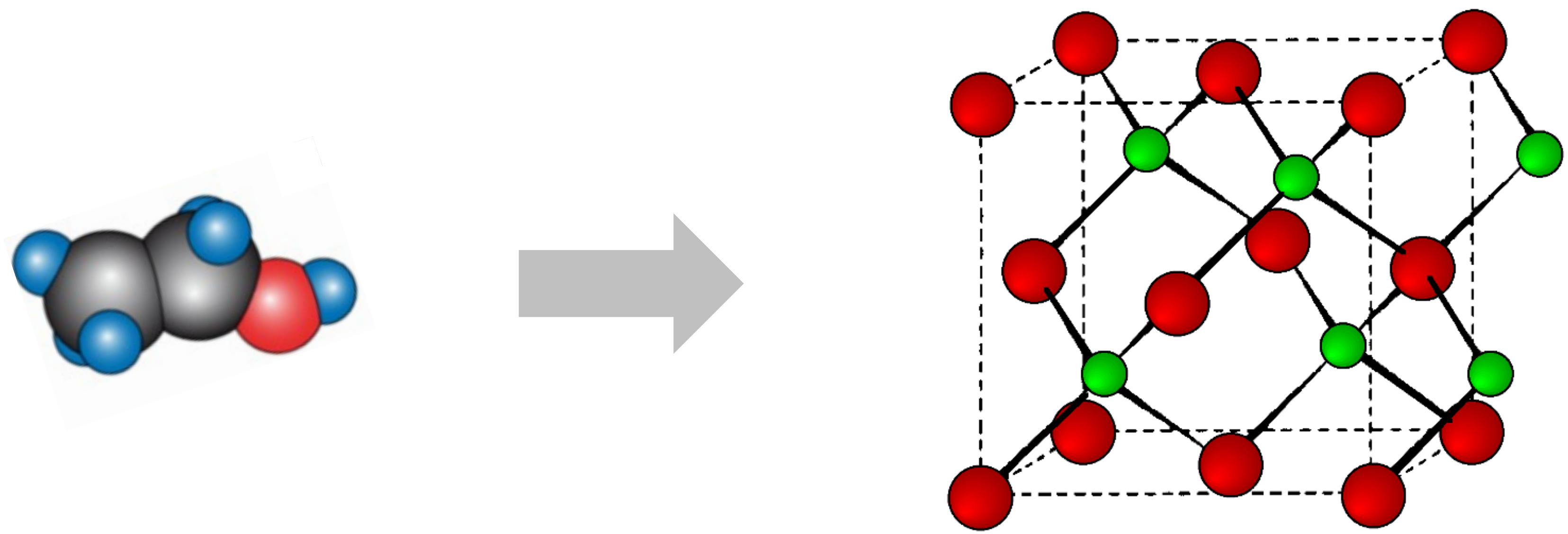
Polarization and magnetization: from molecules to solids



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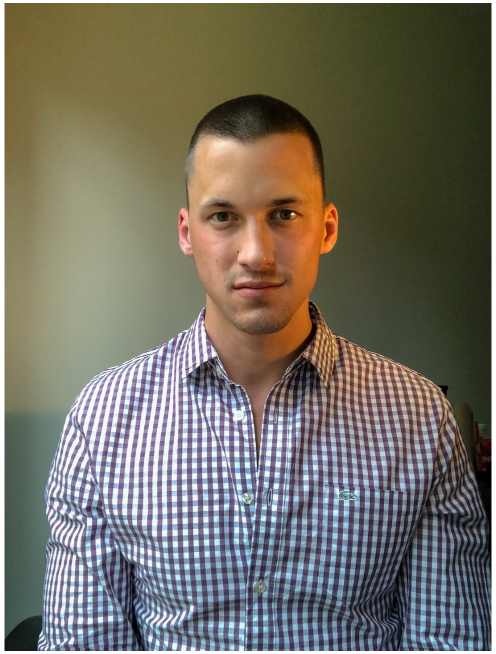
Perry Mahon


Jason Kattan


Rodrigo Muniz


Sylvia Swiecicki


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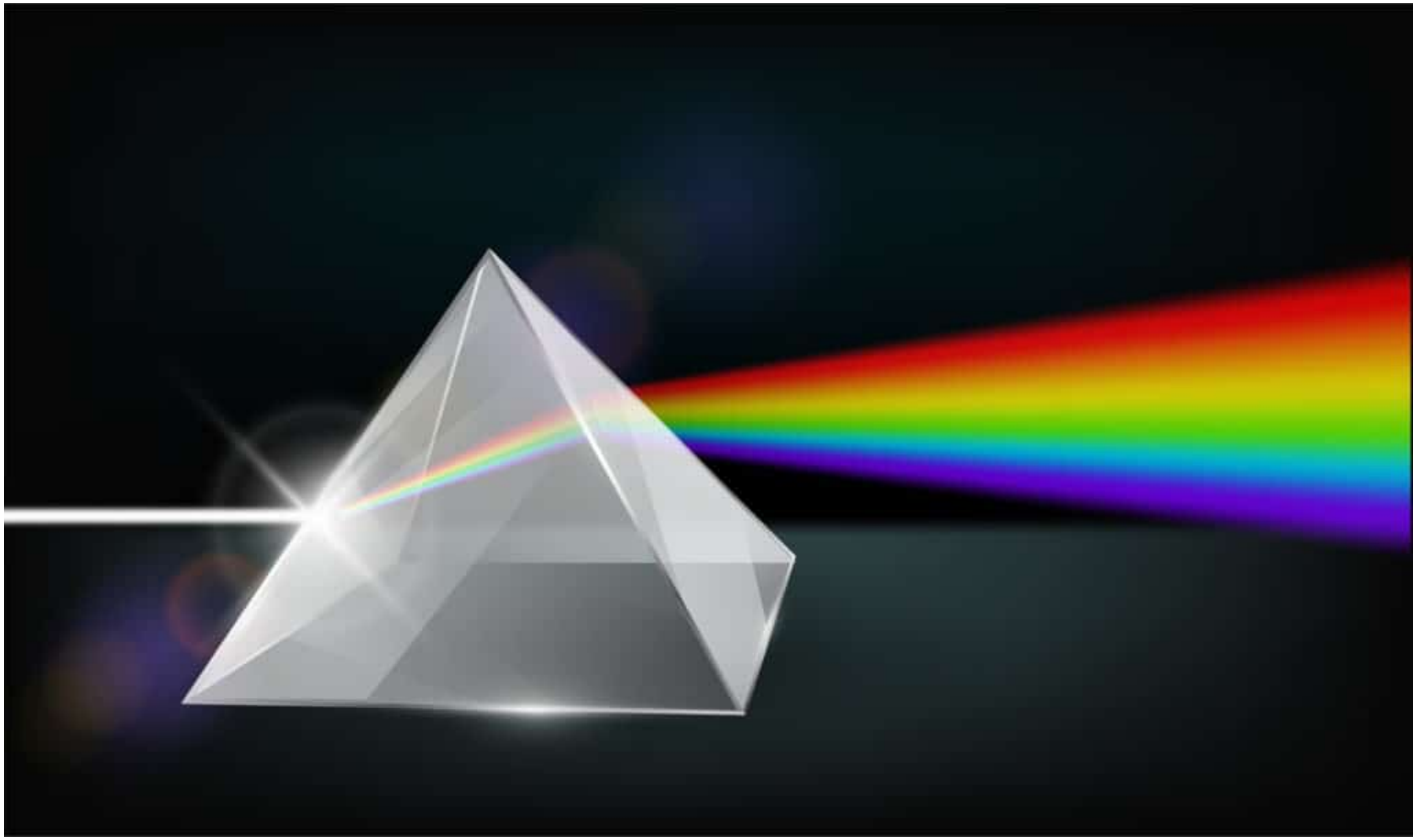
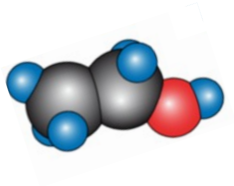
Natural Sciences and Engineering
Research Council of Canada

Overview
The story for molecules and atoms
Generalizing to condensed matter
Some results
Perspective



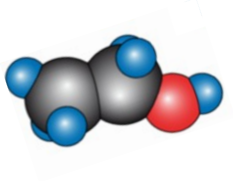


Consider microscopic charge and current densities

$$\rho(\boldsymbol{x}, t) \qquad \boldsymbol{j}(\boldsymbol{x}, t)$$

subject to time-dependent electromagnetic fields

$$\boldsymbol{e}(\boldsymbol{x}, t) \qquad \boldsymbol{b}(\boldsymbol{x}, t)$$



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$$\rho(\boldsymbol{x}, t) \qquad \boldsymbol{j}(\boldsymbol{x}, t)$$

subject to time-dependent electromagnetic fields

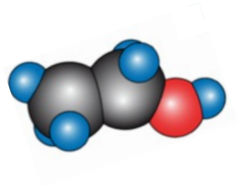
$$\boldsymbol{e}(\boldsymbol{x}, t) \qquad \boldsymbol{b}(\boldsymbol{x}, t)$$

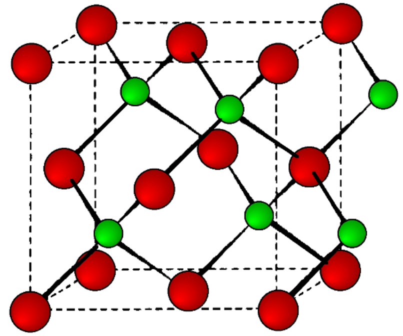
Introduce microscopic polarization and magnetization fields

$$\boldsymbol{p}(\boldsymbol{x}, t) \qquad \boldsymbol{m}(\boldsymbol{x}, t)$$

$$\rho = -\nabla \cdot \boldsymbol{p}$$

$$\boldsymbol{j} = \frac{\partial \boldsymbol{p}}{\partial t} + c\nabla \times \boldsymbol{m}$$





Consider microscopic charge and current densities

$$\rho(\boldsymbol{x},t) \qquad \boldsymbol{j}(\boldsymbol{x},t)$$

subject to time-dependent electromagnetic fields

$$\boldsymbol{e}(\boldsymbol{x},t) \qquad \boldsymbol{b}(\boldsymbol{x},t)$$

Introduce microscopic polarization and magnetization fields (and free charge and current densities)

$$\rho_{free}(\boldsymbol{x},t) \qquad \boldsymbol{j}_{free}(\boldsymbol{x},t)$$

$$\rho = -\nabla\cdot\boldsymbol{p}$$

$$\boldsymbol{j} = \frac{\partial \boldsymbol{p}}{\partial t} + c\nabla\times\boldsymbol{m}$$

$$\rho = -\nabla\cdot\boldsymbol{p} + \rho_{free}$$

$$\boldsymbol{j} = \frac{\partial \boldsymbol{p}}{\partial t} + c\nabla\times\boldsymbol{m} + \boldsymbol{j}_{free}$$

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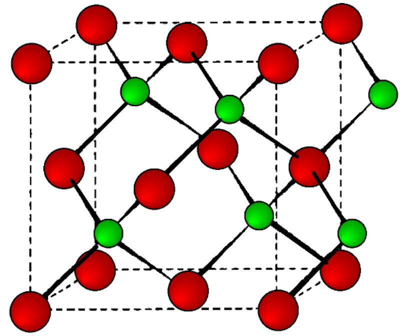

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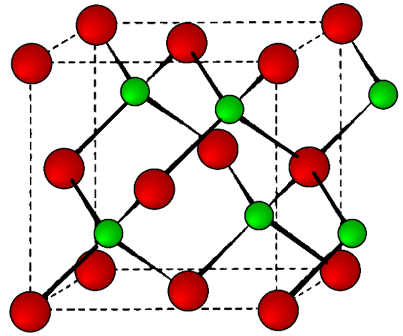

spatially average

$$\varrho = -\nabla \cdot \boldsymbol{P} + \varrho_{free}$$

$$\boldsymbol{J} = \frac{\partial \boldsymbol{P}}{\partial t} + c\nabla \times \boldsymbol{M} + \boldsymbol{J}_{free}$$

$$\rho = -\nabla \cdot \boldsymbol{p} + \rho_{free}$$

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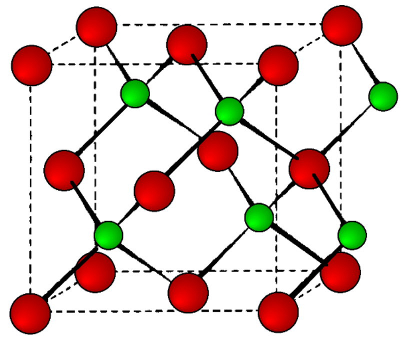

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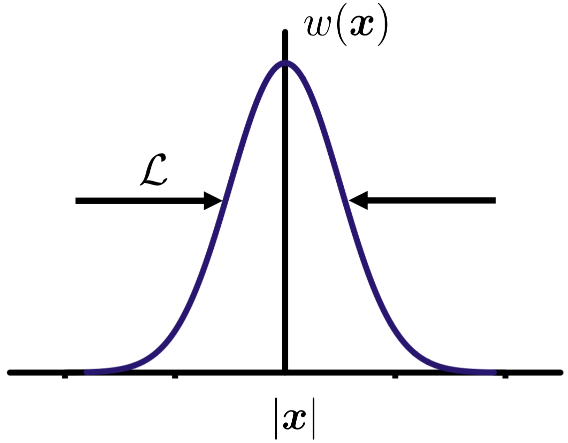
$$\boldsymbol{J} = \frac{\partial \boldsymbol{P}}{\partial t} + c\nabla \times \boldsymbol{M} + \boldsymbol{J}_{free}$$

$$\boldsymbol{P}(\boldsymbol{x}, t) = \int w(\boldsymbol{x} - \boldsymbol{x}')\boldsymbol{p}(\boldsymbol{x}', t)d\boldsymbol{x}'$$

lattice spacing $\ll \mathcal{L} \ll$ *wavelength of light*



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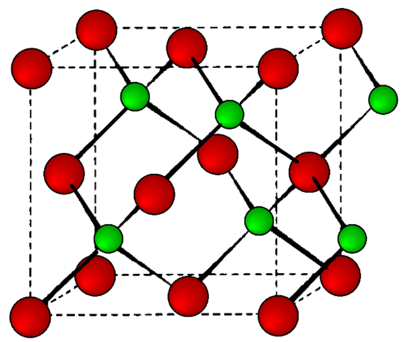

spatially average

$$\varrho = -\nabla \cdot \boldsymbol{P} + \varrho_{free}$$

$$\boldsymbol{J} = \frac{\partial \boldsymbol{P}}{\partial t} + c\nabla \times \boldsymbol{M} + \boldsymbol{J}_{free}$$

$$\nabla \cdot \boldsymbol{e} = 4\pi\rho$$

$$\nabla \cdot \boldsymbol{b} = 0$$

$$c\nabla \times \boldsymbol{b} = 4\pi\boldsymbol{j} + \frac{\partial \boldsymbol{e}}{\partial t}$$

$$c\nabla \times \boldsymbol{e} + \frac{\partial \boldsymbol{b}}{\partial t} = 0$$

spatially average

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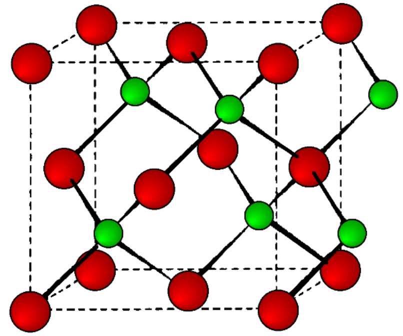

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spatially average

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$$c\nabla \times \boldsymbol{b} = 4\pi\boldsymbol{j} + \frac{\partial \boldsymbol{e}}{\partial t}$$

$$c\nabla \times \boldsymbol{e} + \frac{\partial \boldsymbol{b}}{\partial t} = 0$$

$$\boldsymbol{D} = \boldsymbol{E} + 4\pi\boldsymbol{P}$$

$$\boldsymbol{H} = \boldsymbol{B} - 4\pi\boldsymbol{M}$$

spatially average

$$\nabla \cdot \boldsymbol{E} = 4\pi\varrho$$

$$\nabla \cdot \boldsymbol{B} = 0$$

$$c\nabla \times \boldsymbol{B} = 4\pi\boldsymbol{J} + \frac{\partial \boldsymbol{E}}{\partial t}$$

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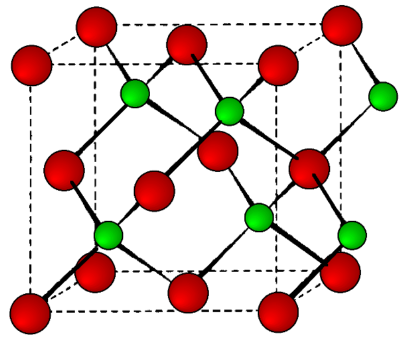


spatially average

$$\varrho = -\nabla \cdot \boldsymbol{P} + \varrho_{free}$$

$$\boldsymbol{J} = \frac{\partial \boldsymbol{P}}{\partial t} + c\nabla \times \boldsymbol{M} + \boldsymbol{J}_{free}$$

$$\boldsymbol{D} = \boldsymbol{E} + 4\pi \boldsymbol{P}$$

$$\boldsymbol{H} = \boldsymbol{B} - 4\pi \boldsymbol{M}$$

$$\nabla \cdot \boldsymbol{D} = 4\pi \varrho_{free}$$

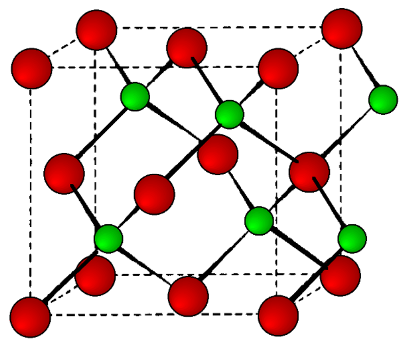
$$\nabla \cdot \boldsymbol{B} = 0$$

$$c\nabla \times \boldsymbol{H} = 4\pi \boldsymbol{J}_{free} + \frac{\partial \boldsymbol{D}}{\partial t}$$

$$c\nabla \times \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = 0$$

$$\rho = -\nabla \cdot \boldsymbol{p} + \rho_{free}$$

$$\boldsymbol{j} = \frac{\partial \boldsymbol{p}}{\partial t} + c\nabla \times \boldsymbol{m} + \boldsymbol{j}_{free}$$



spatially average

$$\varrho = -\nabla \cdot \boldsymbol{P} + \varrho_{free}$$

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can formulate the dynamics of

$\boldsymbol{p}(\boldsymbol{x},t)$ $\boldsymbol{m}(\boldsymbol{x},t)$ $\rho_{free}(\boldsymbol{x},t)$ $\boldsymbol{j}_{free}(\boldsymbol{x},t)$

in terms of

$\boldsymbol{e}(\boldsymbol{x},t)$ $\boldsymbol{b}(\boldsymbol{x},t)$

and then within various approximations susceptibilities can be extracted

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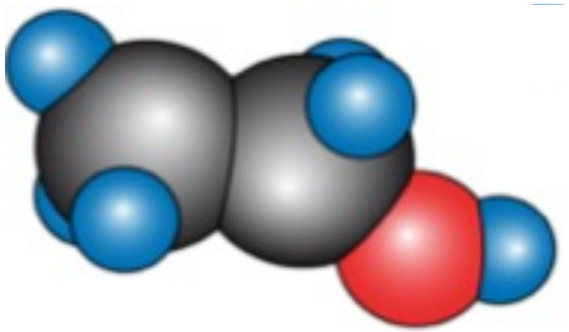
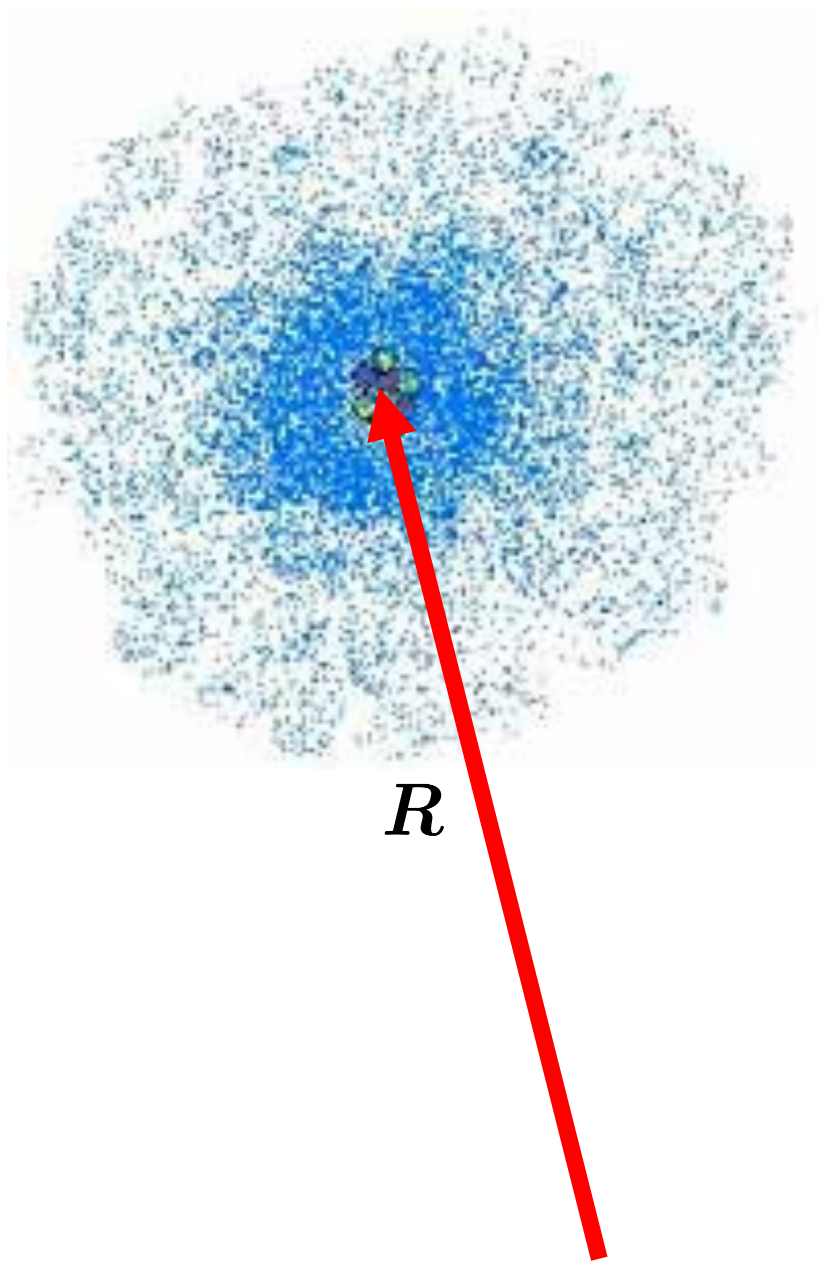


The story for molecules and atoms

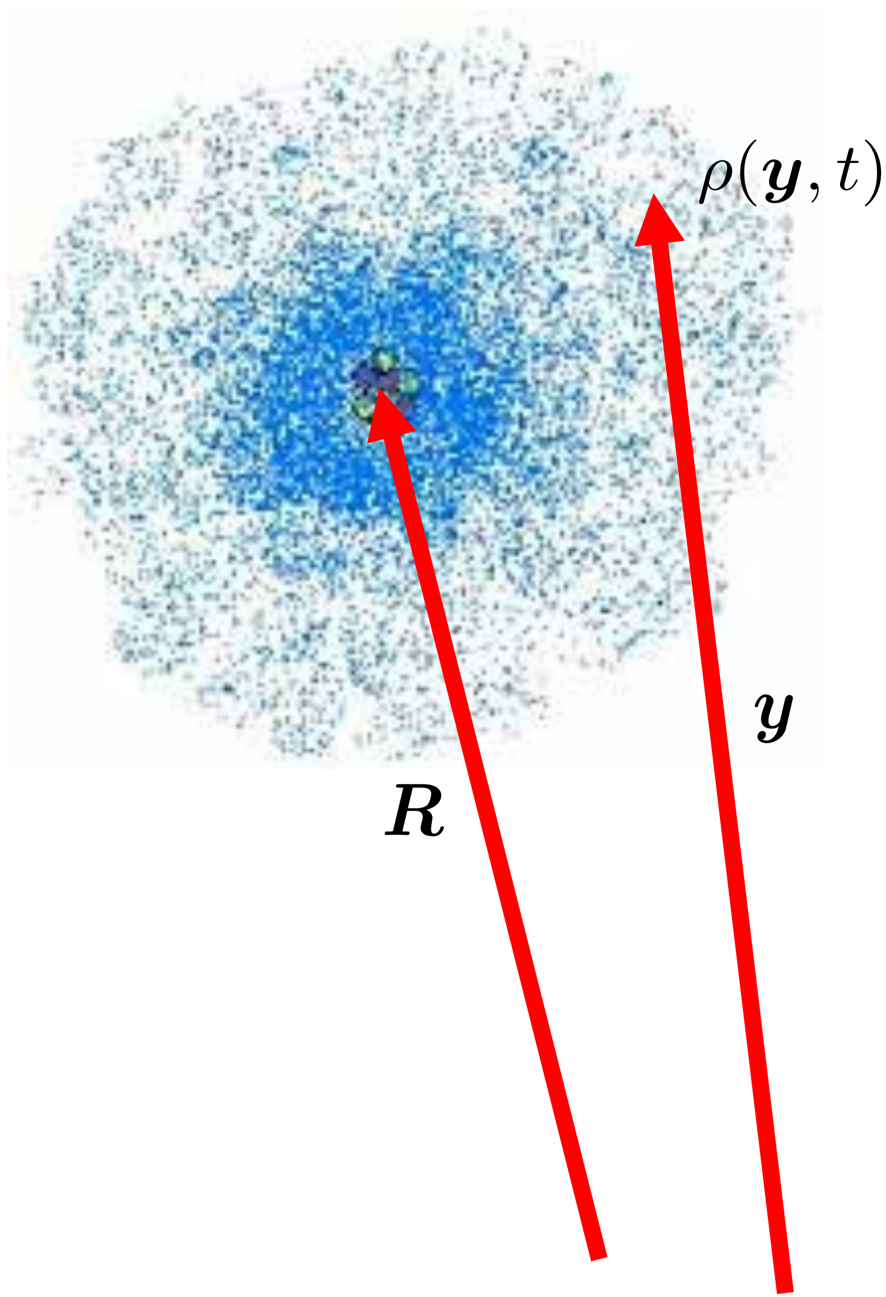
Power,
Zienau,
Wooley,
Thirunamachandran,
Healy...

D.P. Craig and T. Thirunamachandran,
"Molecular quantum electrodynamics," Academic Press
W.P. Healy,
"Nonrelativistic quantum electrodynamics," Academic Press

defining the microscopic polarization $\mathbf{p}(\boldsymbol{x}, t)$



defining the microscopic polarization $\mathbf{p}(\boldsymbol{x}, t)$



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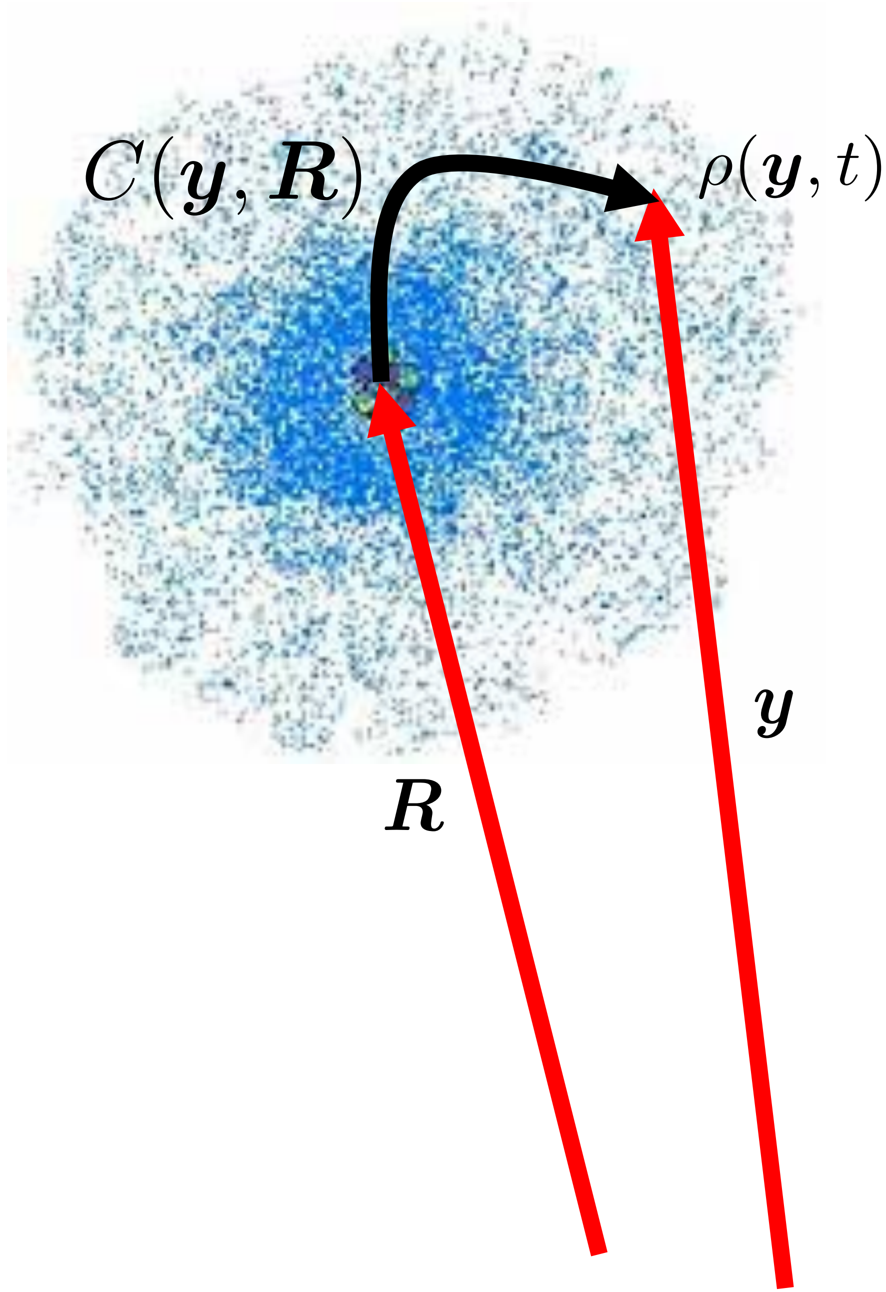

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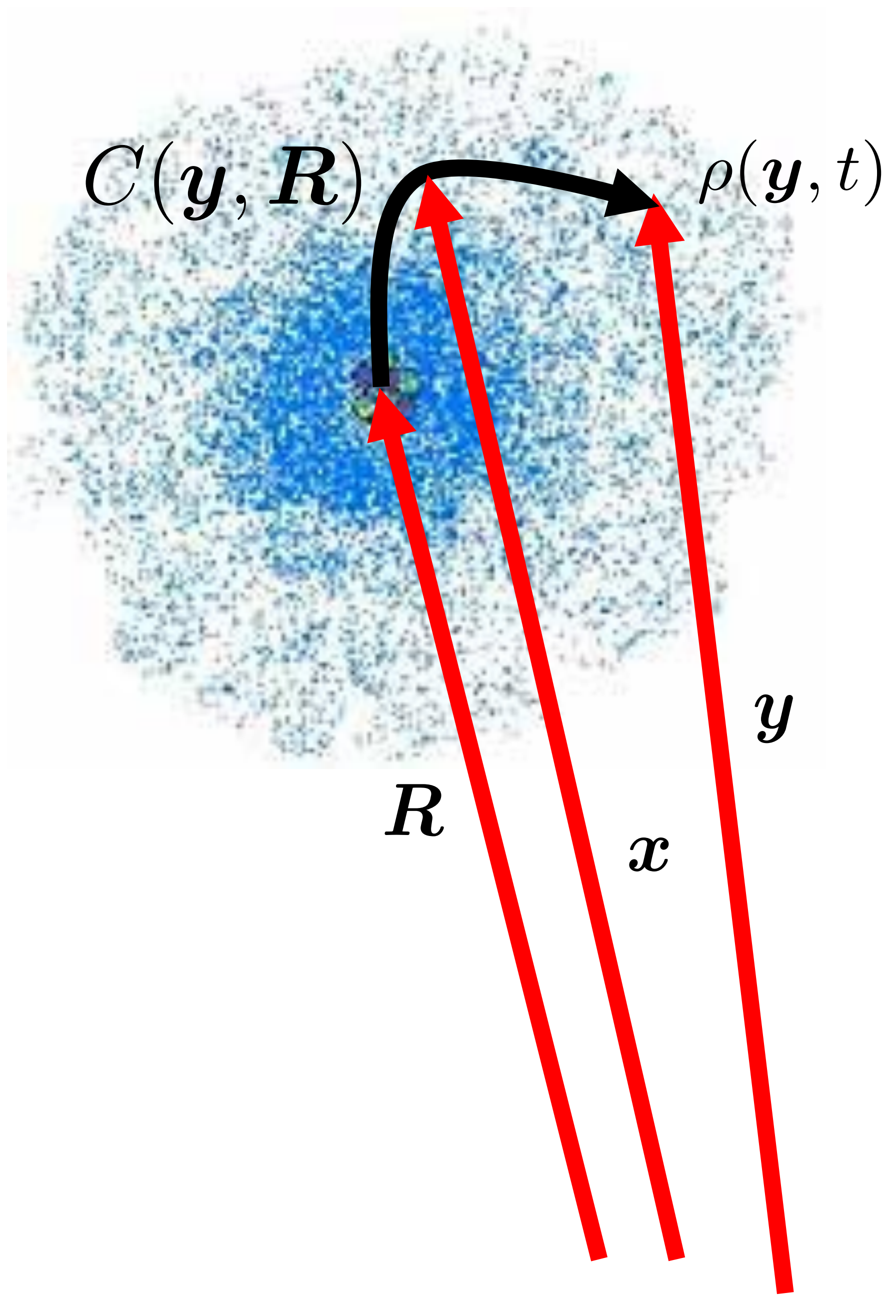

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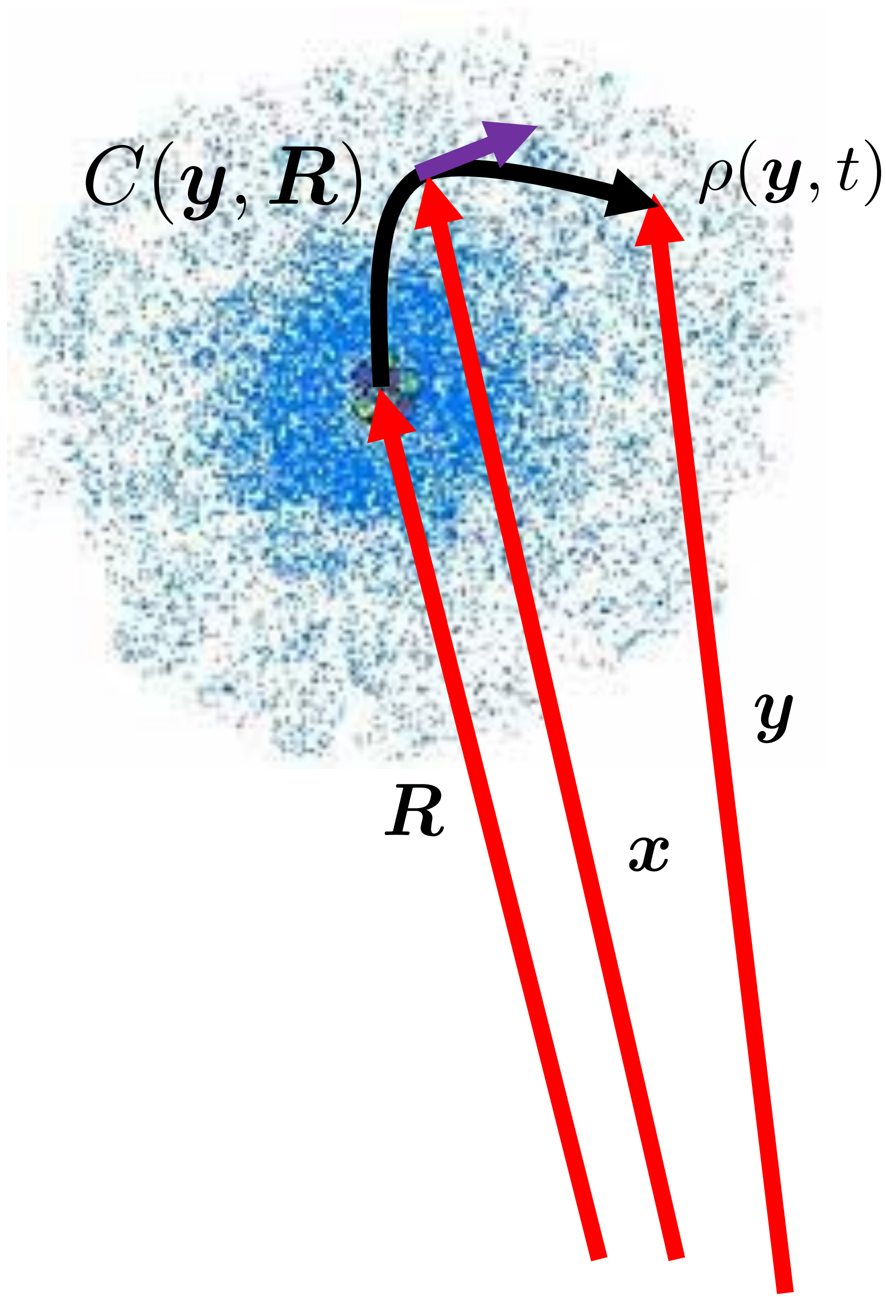

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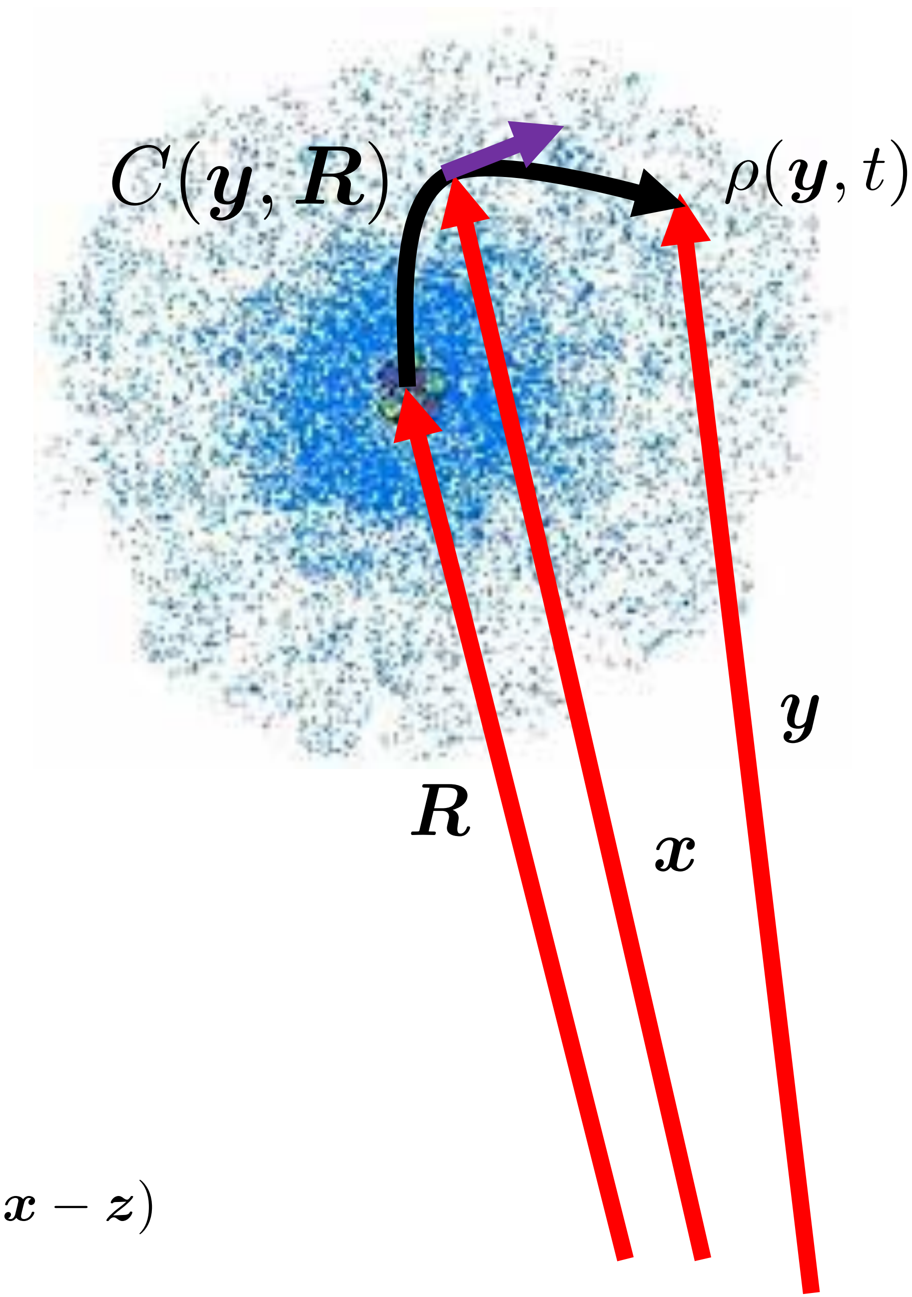
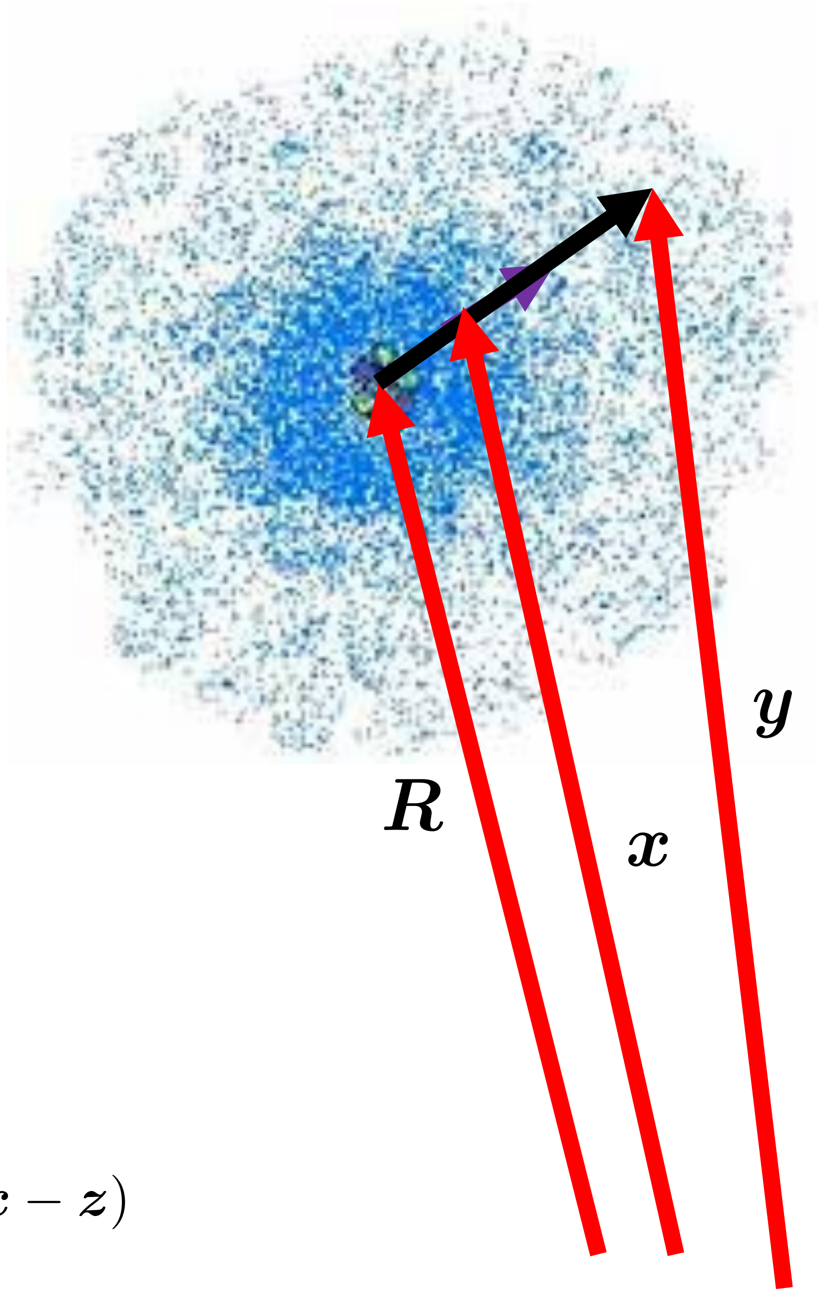


$$\boldsymbol{p}(\boldsymbol{x},t) = \int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho(\boldsymbol{y},t)d\boldsymbol{y}$$

$$\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R}) = \int_{C(\boldsymbol{y},\boldsymbol{R})} d\boldsymbol{z}\delta(\boldsymbol{x}-\boldsymbol{z})$$



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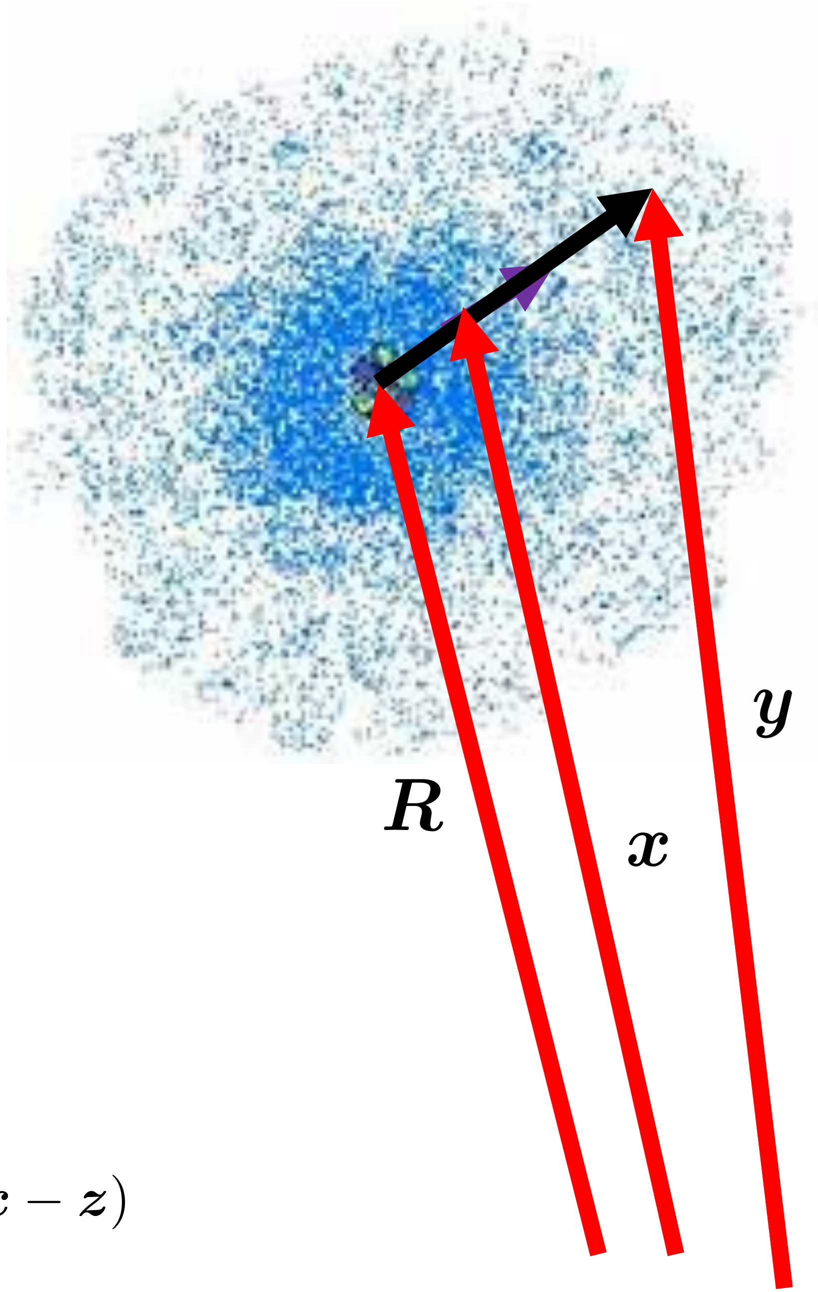
$$\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R}) = \int_{\text{line from } \boldsymbol{R} \text{ to } \boldsymbol{y}} d\boldsymbol{z}\delta(\boldsymbol{x}-\boldsymbol{z})$$

$$\int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})d\boldsymbol{x} = \boldsymbol{y} - \boldsymbol{R}$$

$$\int \boldsymbol{p}(\boldsymbol{x},t)d\boldsymbol{x} = \int (\boldsymbol{y}-\boldsymbol{R})\rho(\boldsymbol{y},t)d\boldsymbol{y}$$

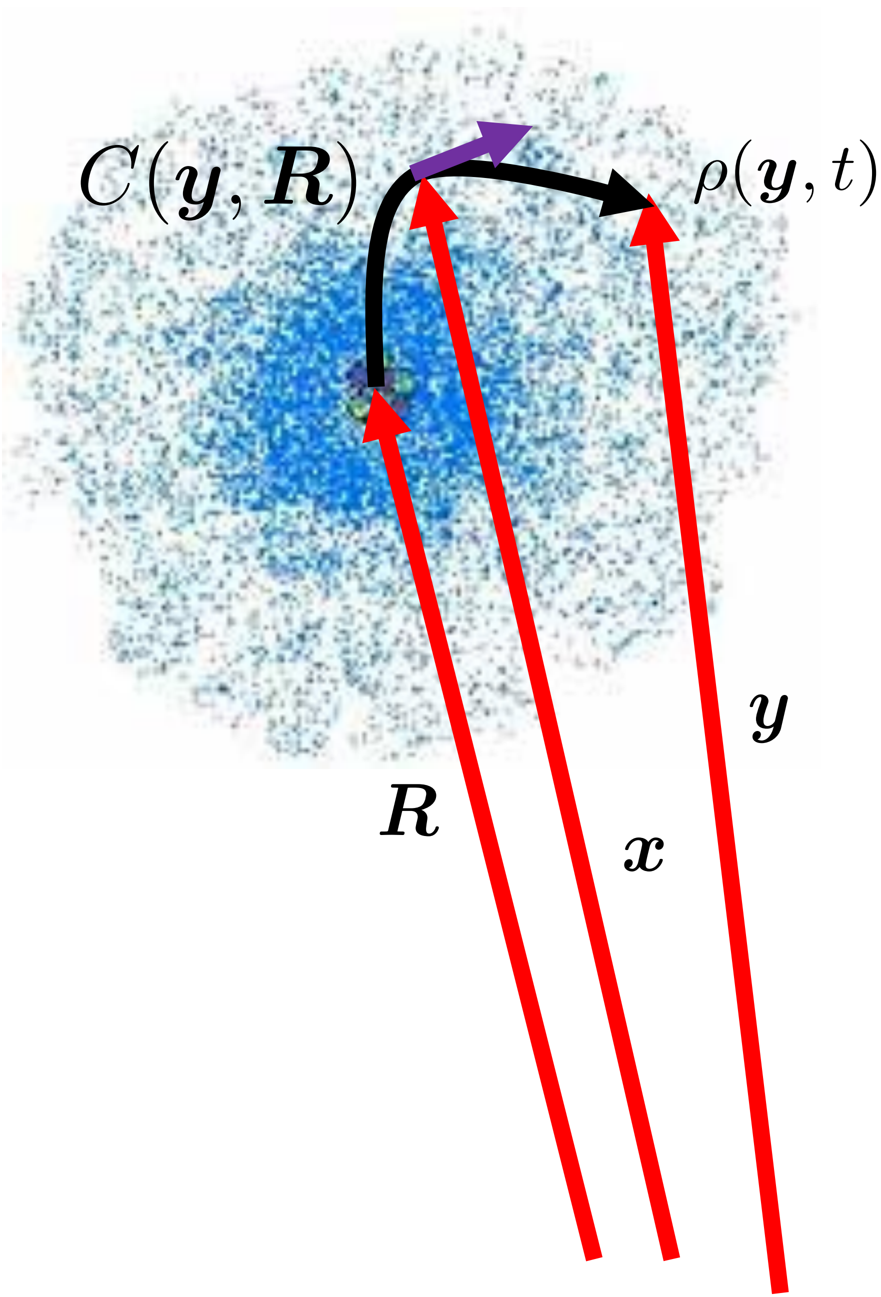
electric dipole moment

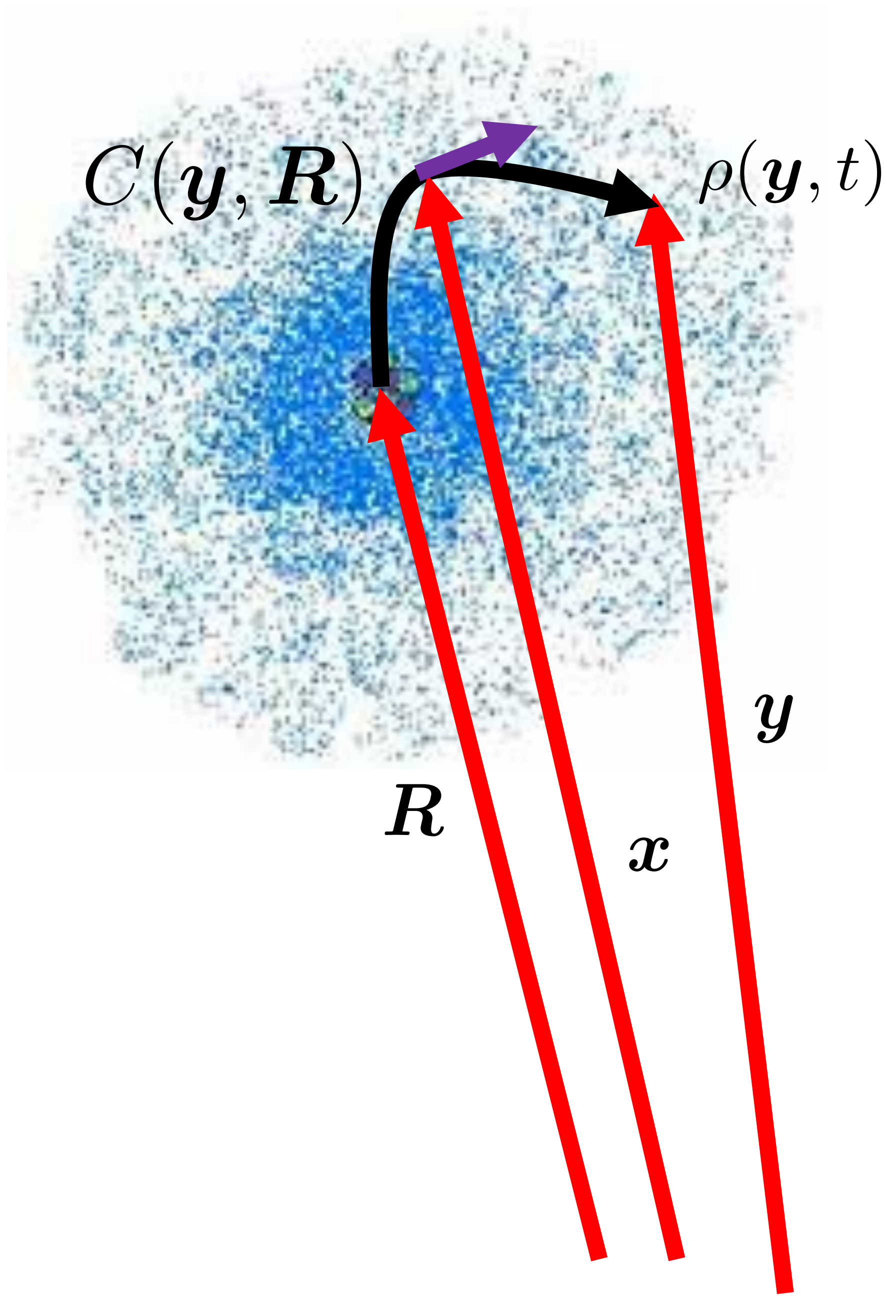


$$\boldsymbol{p}(\boldsymbol{x},t) = \int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho(\boldsymbol{y},t)d\boldsymbol{y}$$

$$\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R}) = \int_{\text{line from } \boldsymbol{R} \text{ to } \boldsymbol{y}} d\boldsymbol{z}\,\delta(\boldsymbol{x}-\boldsymbol{z})$$

$$p^i(\boldsymbol{x},t) = \int s^i(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho(\boldsymbol{y},t)d\boldsymbol{y}$$



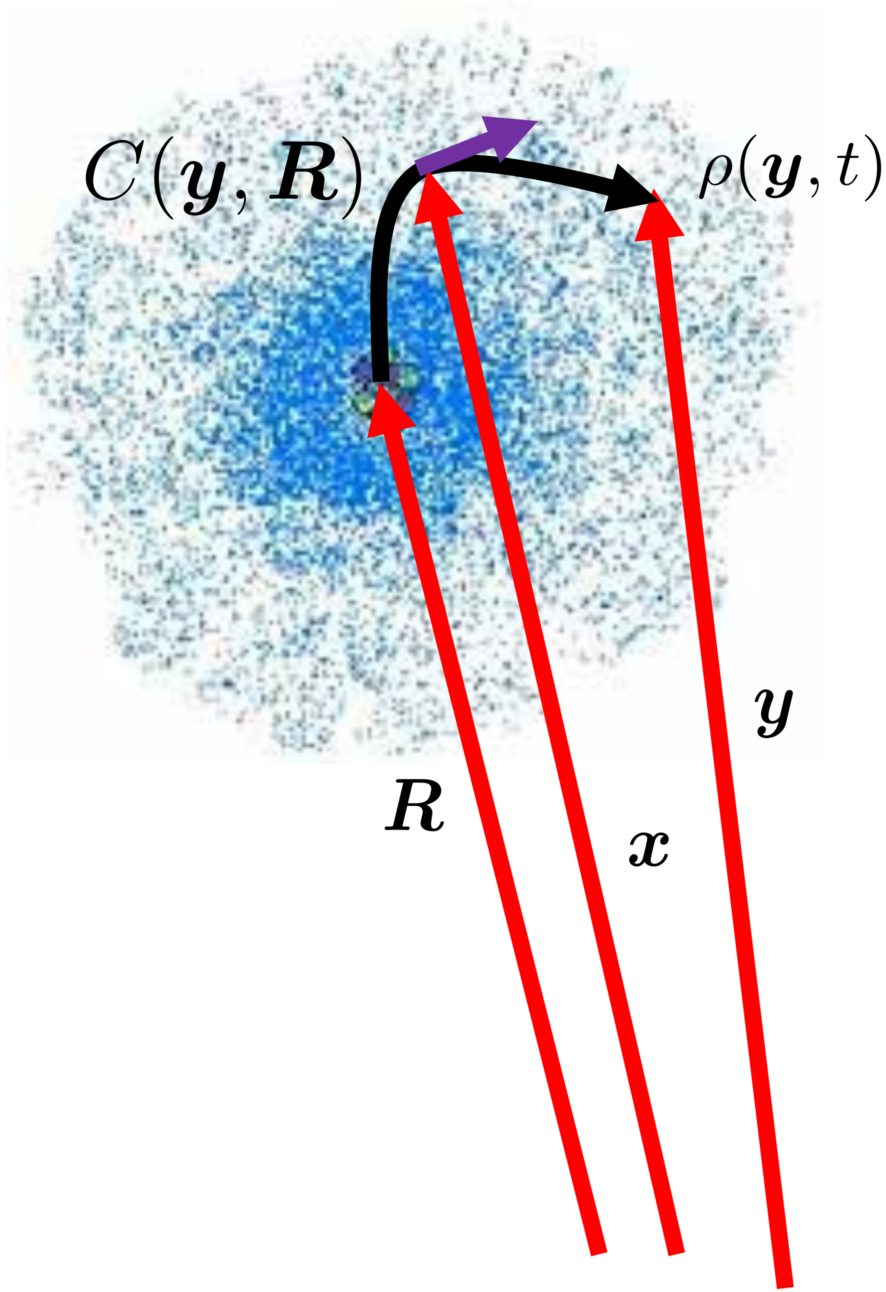


$$p^i(\boldsymbol{x}, t) = \int s^i(\boldsymbol{x}; \boldsymbol{y}, \boldsymbol{R})\rho(\boldsymbol{y}, t)d\boldsymbol{y}$$

$$m^i(\boldsymbol{x}, t) = \frac{1}{c}\int \alpha^{ik}(\boldsymbol{x}; \boldsymbol{y}, \boldsymbol{R})j^k(\boldsymbol{y}, t)d\boldsymbol{y}$$

$$s^i(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R}) = \int_{C(\boldsymbol{y},\boldsymbol{R})} dz^i \delta(\boldsymbol{x}-\boldsymbol{z})$$

$$\alpha^{ik}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R}) = \epsilon^{imn} \int_{C(\boldsymbol{y},\boldsymbol{R})} dz^m \frac{\partial z^n}{\partial x^k} \delta(\boldsymbol{x}-\boldsymbol{z})$$



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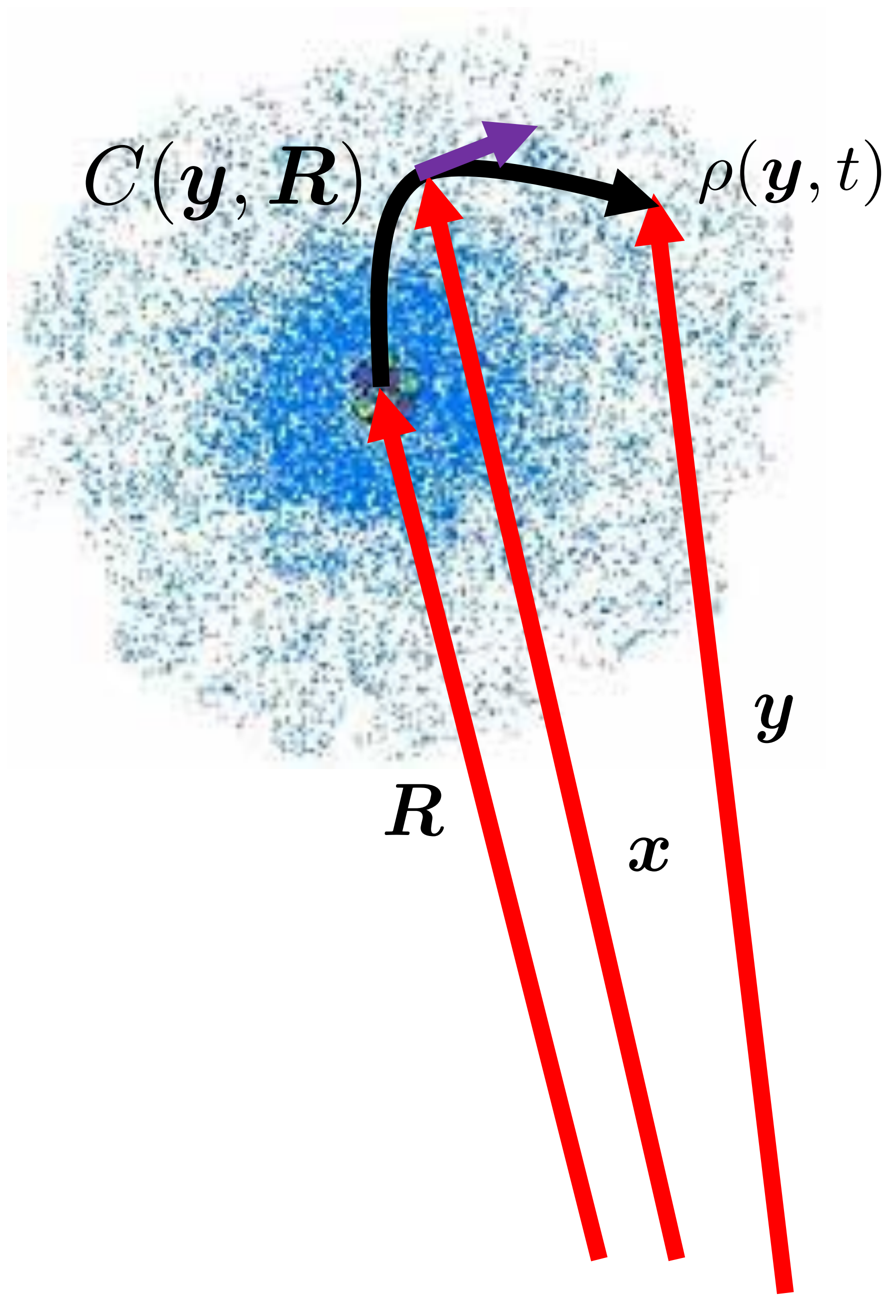
"relators"



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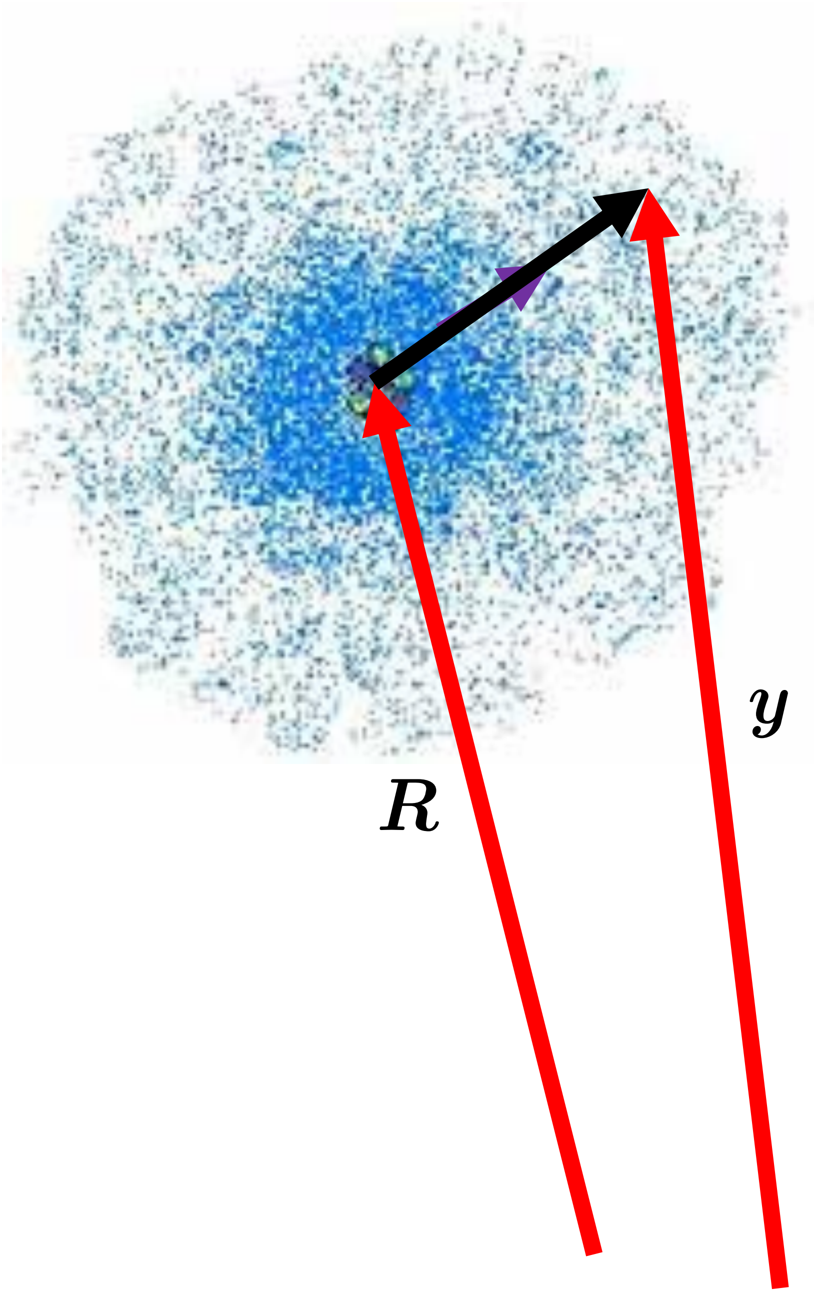

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$$\boldsymbol{j}(\boldsymbol{x},t) = \frac{\partial \boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla \times \boldsymbol{m}(\boldsymbol{x},t)$$

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t)$$

follow since

$$\nabla \cdot \boldsymbol{j}(\boldsymbol{x},t) + \frac{\partial \rho(\boldsymbol{x},t)}{\partial t} = 0$$

$$p^i(\boldsymbol{x},t) = \int s^i(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho(\boldsymbol{y},t)d\boldsymbol{y}$$

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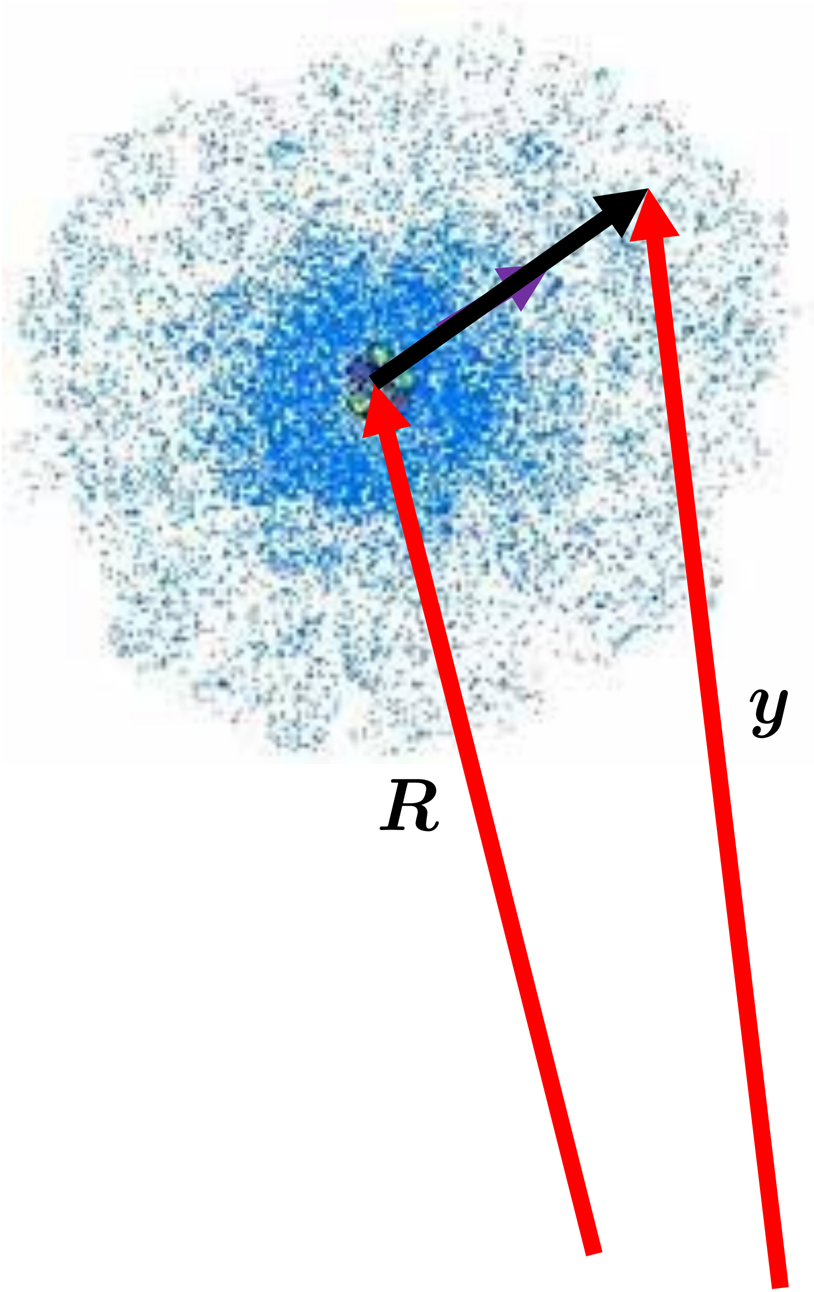
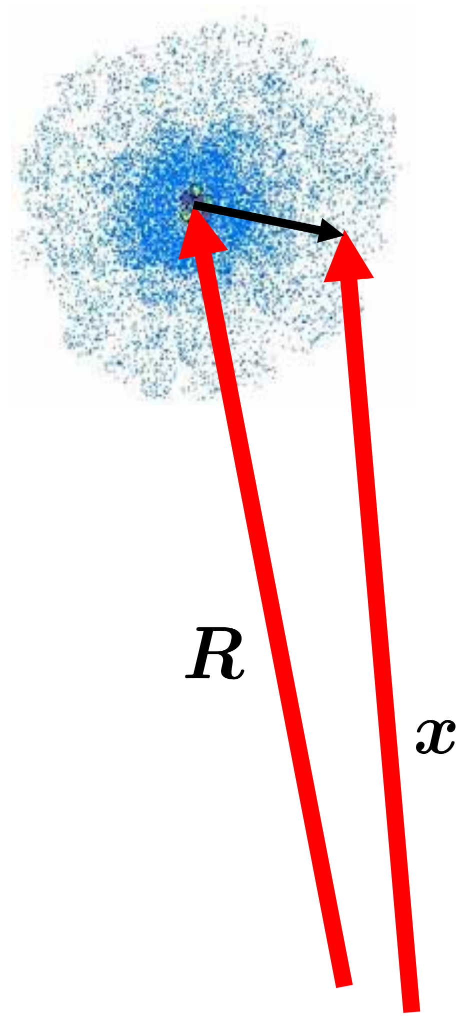


$$\psi_{sp}(\boldsymbol{x},t) = e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi(\boldsymbol{x},t)$$

$$\Phi(\boldsymbol{x},\boldsymbol{R};t) = \frac{e}{\hbar c}\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})A^i(\boldsymbol{w},t)d\boldsymbol{w}$$

generalized Peierls phase



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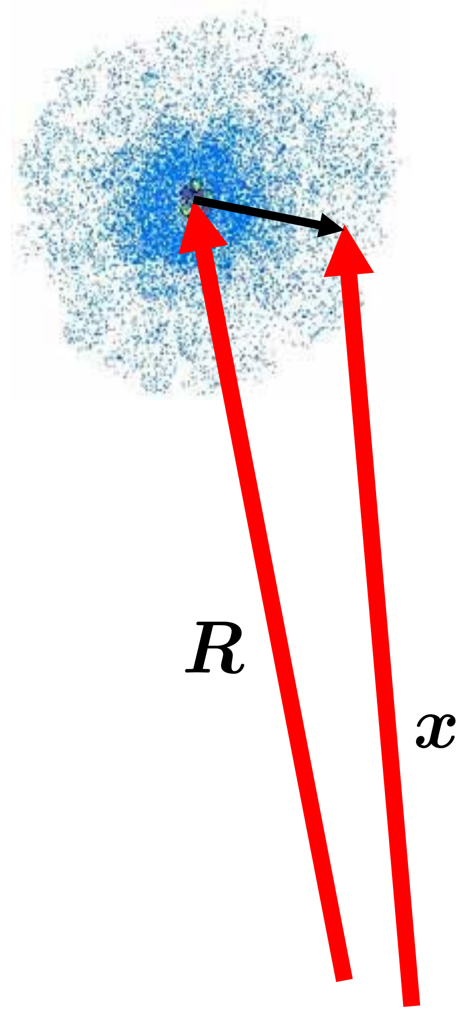
generalized Peierls phase

Wilson line integral



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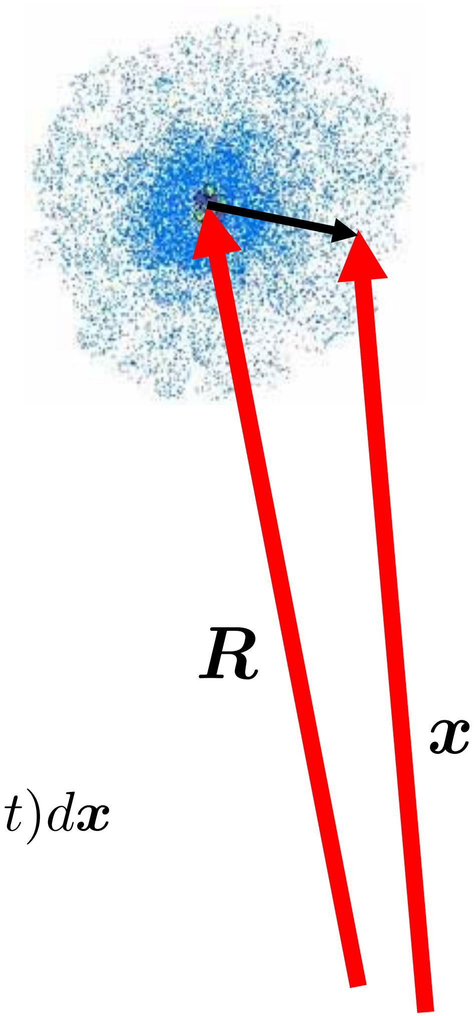
generalized Peierls phase



$$H_{sp}(t) = H_0 - \int \boldsymbol{p}(\boldsymbol{x},t)\cdot\boldsymbol{E}(\boldsymbol{x},t)d\boldsymbol{x}$$
$$- \int \boldsymbol{m}_P(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x} - \frac{1}{2}\int \boldsymbol{m}_D(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x}$$

$$\psi_{sp}(\boldsymbol{x},t) = e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi(\boldsymbol{x},t)$$

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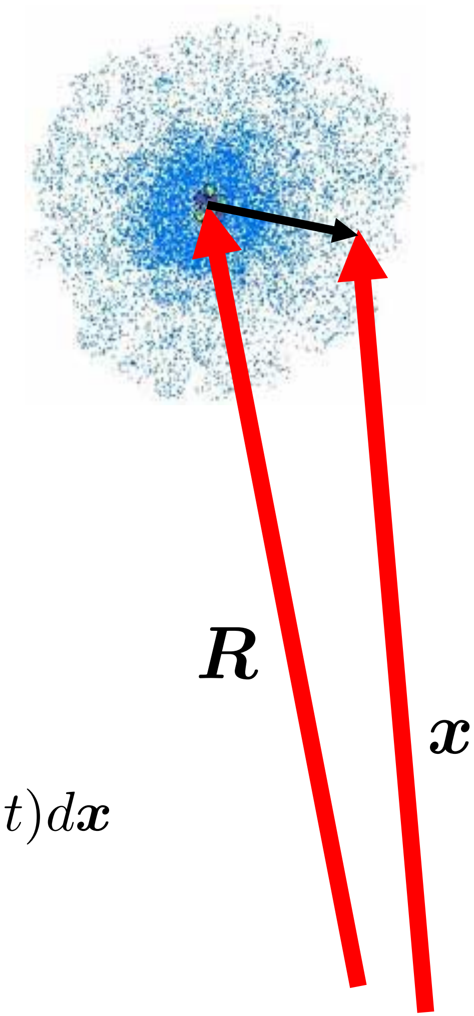
generalized Peierls phase



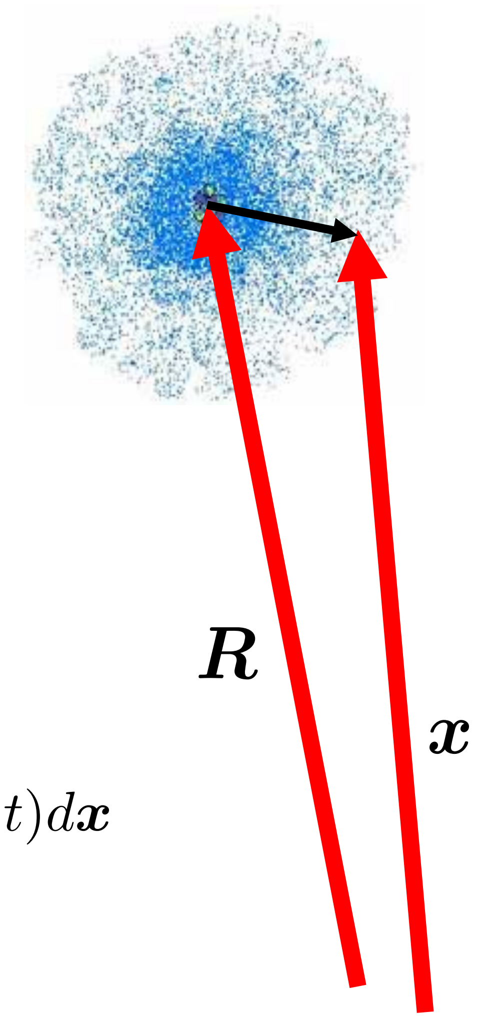
$$H_{sp}(t) = H_0 - \int \boldsymbol{p}(\boldsymbol{x},t)\cdot\boldsymbol{E}(\boldsymbol{x},t)d\boldsymbol{x} - \int \boldsymbol{m}_P(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x} - \frac{1}{2}\int \boldsymbol{m}_D(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x}$$

Gauge freedom of electromagnetic field replaced by "path freedom" of relators

$$\psi_{sp}(\boldsymbol{x},t) = e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi(\boldsymbol{x},t)$$

$$\Phi(\boldsymbol{x},\boldsymbol{R};t) = \frac{e}{\hbar c}\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})A^i(\boldsymbol{w},t)d\boldsymbol{w}$$

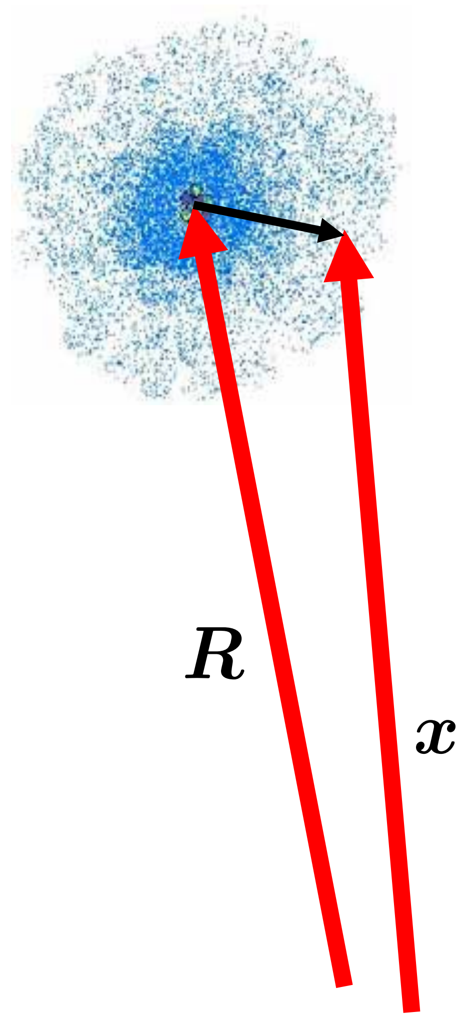
generalized Peierls phase



$$H_{sp}(t) = H_0 - \int \boldsymbol{p}(\boldsymbol{x},t)\cdot\boldsymbol{E}(\boldsymbol{x},t)d\boldsymbol{x}$$
$$- \int \boldsymbol{m}_P(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x} - \frac{1}{2}\int \boldsymbol{m}_D(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x}$$

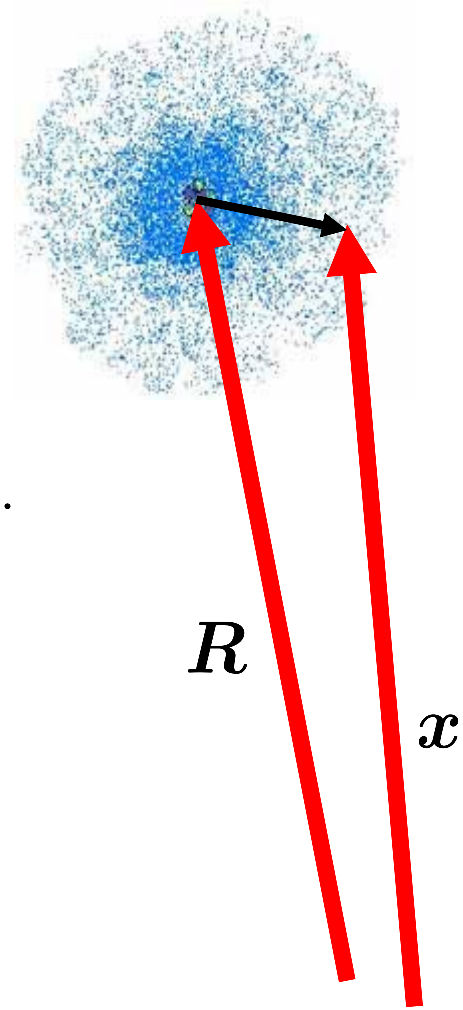
Gauge freedom of electromagnetic field replaced by "path freedom" of relators

$$\int p^i(\boldsymbol{x},t)E^i(\boldsymbol{x},t)d\boldsymbol{x}$$

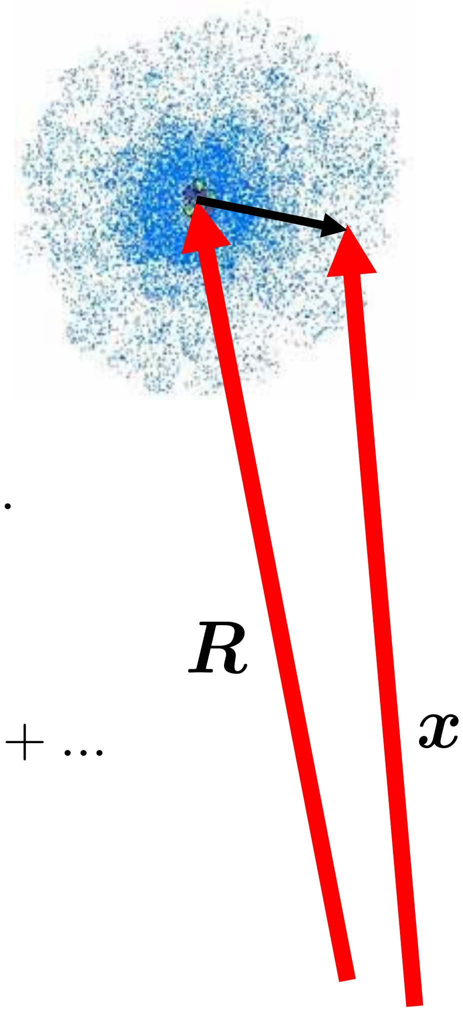


$$\int p^i(\boldsymbol{x},t)E^i(\boldsymbol{x},t)d\boldsymbol{x}$$

$$E^i(\boldsymbol{x},t) = E^i(\boldsymbol{R},t) + \frac{\partial E^i(\boldsymbol{R},t)}{\partial R^j}(x^j - R^j) + ...$$



$$\int p^i(\boldsymbol{x},t)E^i(\boldsymbol{x},t)d\boldsymbol{x}$$



$$E^i(\boldsymbol{x},t) = E^i(\boldsymbol{R},t) + \frac{\partial E^i(\boldsymbol{R},t)}{\partial R^j}(x^j - R^j) + ...$$

$$\int p^i(\boldsymbol{x},t)E^i(\boldsymbol{x},t)d\boldsymbol{x} \;\Rightarrow\; \mu^i E^i(\boldsymbol{R},t) + q^{ij}\frac{\partial E^i(\boldsymbol{R},t)}{\partial R^j} + ...$$

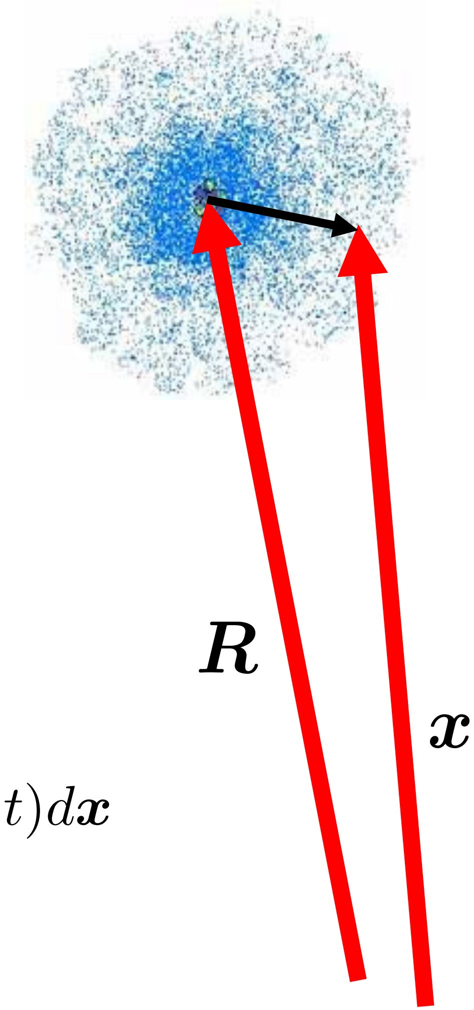
$$\mu^i = \int (y^i - R^i)\rho(\boldsymbol{y},t)d\boldsymbol{y}$$ ***electric dipole moment***

$$q^{ij} = \frac{1}{2}\int (y^i - R^i)(y^j - R^j)\rho(\boldsymbol{y},t)d\boldsymbol{y}$$ ***electric quadrupole moment***

$$\psi_{sp}(\boldsymbol{x},t) = e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi(\boldsymbol{x},t)$$

$$\Phi(\boldsymbol{x},\boldsymbol{R};t) = \frac{e}{\hbar c}\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})A^i(\boldsymbol{w},t)d\boldsymbol{w}$$

generalized Peierls phase



$$H_{sp}(t) = H_0 - \int \boldsymbol{p}(\boldsymbol{x},t)\cdot\boldsymbol{E}(\boldsymbol{x},t)d\boldsymbol{x} - \int \boldsymbol{m}_P(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x} - \frac{1}{2}\int \boldsymbol{m}_D(\boldsymbol{x},t)\cdot\boldsymbol{B}(\boldsymbol{x},t)d\boldsymbol{x}$$

electric dipole moment operator

paramagnetic dipole moment operator

$$\Rightarrow \quad H_0 - \boldsymbol{\mu}\cdot\boldsymbol{E}(\boldsymbol{R},t) - \boldsymbol{\nu}_P\cdot\boldsymbol{B}(\boldsymbol{R},t) - \frac{1}{2}\boldsymbol{\nu}_D\cdot\boldsymbol{B}(\boldsymbol{R},t) + \cdots$$

diamagnetic dipole moment operator

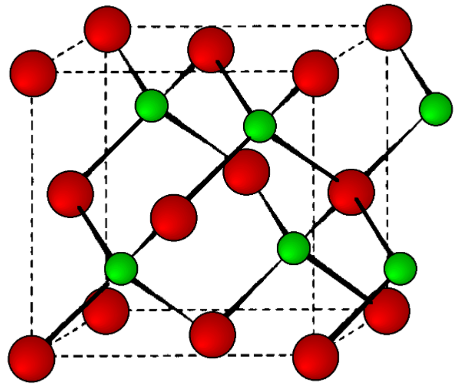
Overview

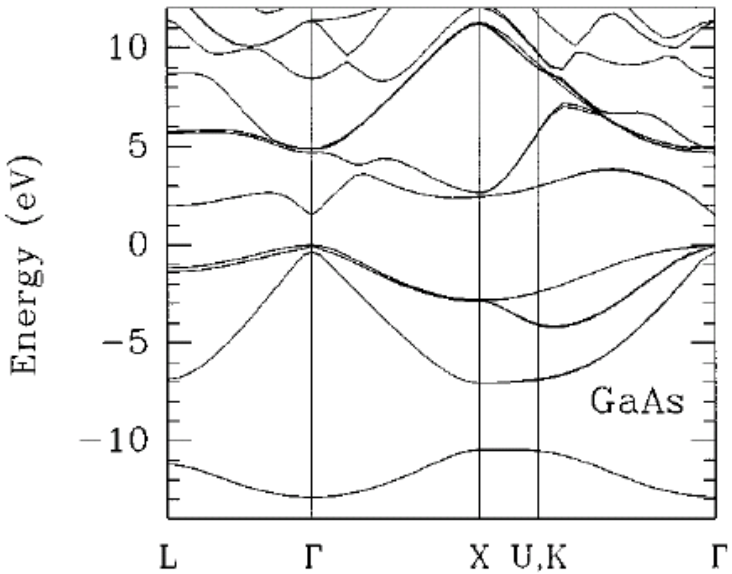
The story for molecules and atoms

Generalizing to condensed matter

Some results

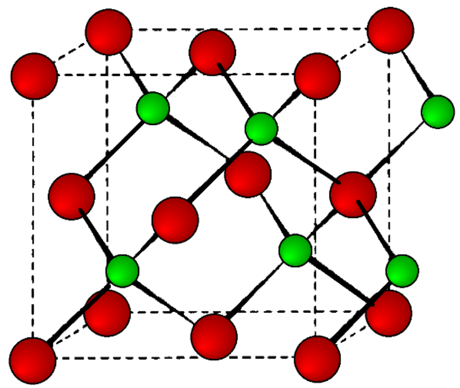
Perspective

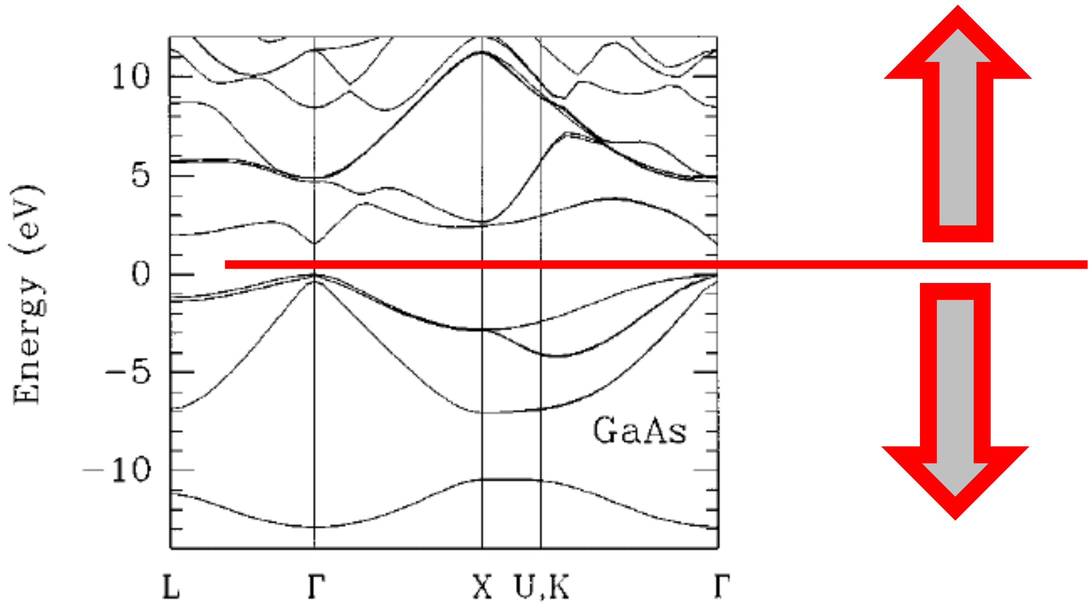




Bloch functions

$$\phi_{n\boldsymbol{k}}(\boldsymbol{x}) = \frac{u_{n\boldsymbol{k}}(\boldsymbol{x})}{(2\pi)^{3/2}} e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$$



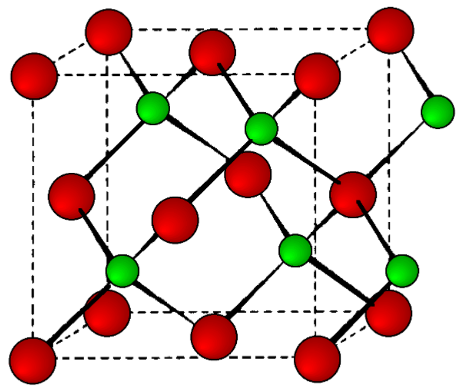


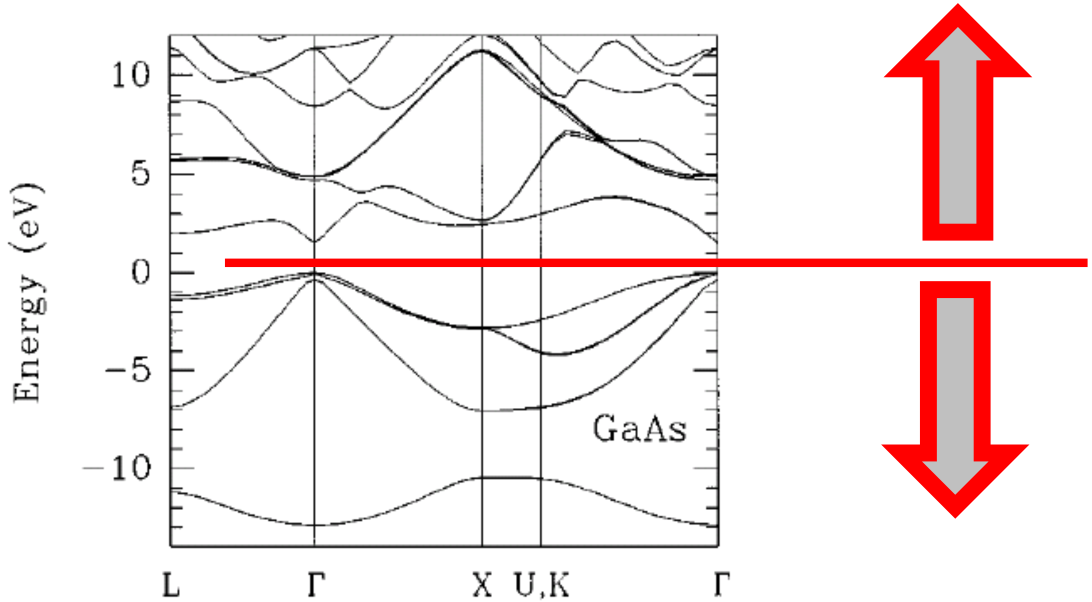
empty conduction bands

filled valence bands

Bloch functions

$$\phi_{n\boldsymbol{k}}(\boldsymbol{x}) = \frac{u_{n\boldsymbol{k}}(\boldsymbol{x})}{(2\pi)^{3/2}} e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$$





empty conduction bands

filled valence bands

construct exponentially localized Wannier functions

Bloch functions

$$\phi_{n\boldsymbol{k}}(\boldsymbol{x}) = \frac{u_{n\boldsymbol{k}}(\boldsymbol{x})}{(2\pi)^{3/2}} e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$$

$$W_{\alpha \boldsymbol{R}}(\boldsymbol{x}) = \sqrt{\Omega_{uc}} \int_{BZ} \frac{d\boldsymbol{k}}{(2\pi)^{3/2}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}} \sum_n \phi_{n\boldsymbol{k}}(\boldsymbol{x}) U_{n\alpha}(\boldsymbol{k})$$

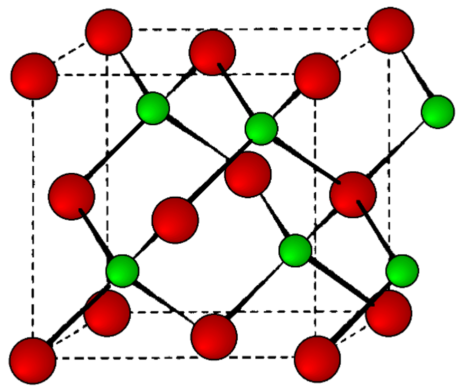
valence Wannier function type

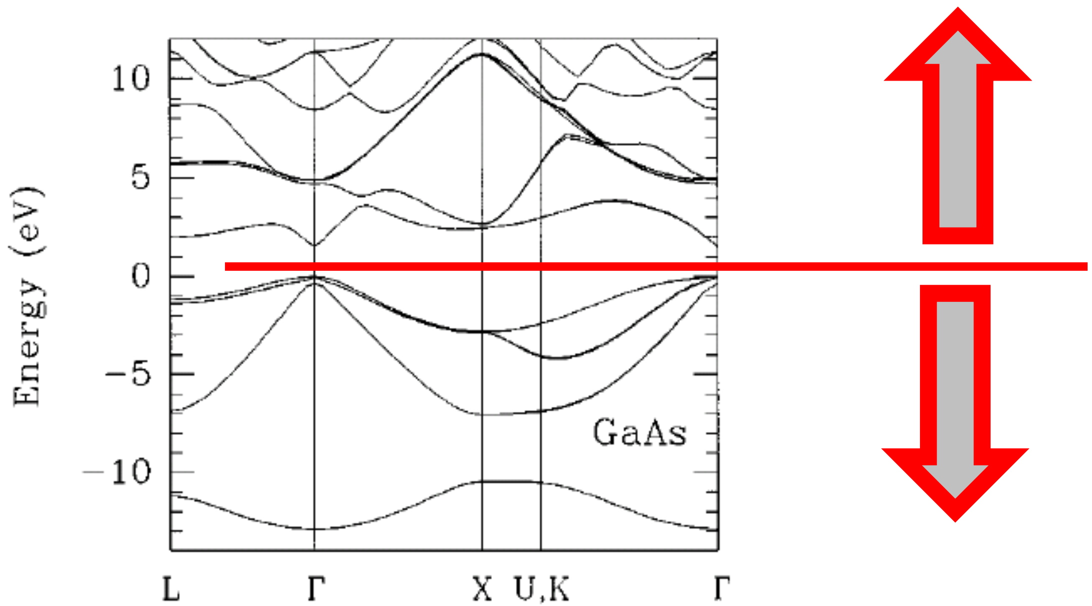
lattice site

unit cell volume

valence bands

unitary matrix





empty conduction bands

filled valence bands

construct exponentially localized Wannier functions

Bloch functions

$$\phi_{n\boldsymbol{k}}(\boldsymbol{x}) = \frac{u_{n\boldsymbol{k}}(\boldsymbol{x})}{(2\pi)^{3/2}} e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$$

$$W_{\alpha \boldsymbol{R}}(\boldsymbol{x}) = \sqrt{\Omega_{uc}} \int_{BZ} \frac{d\boldsymbol{k}}{(2\pi)^{3/2}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}} \sum_n \phi_{n\boldsymbol{k}}(\boldsymbol{x}) U_{n\alpha}(\boldsymbol{k})$$

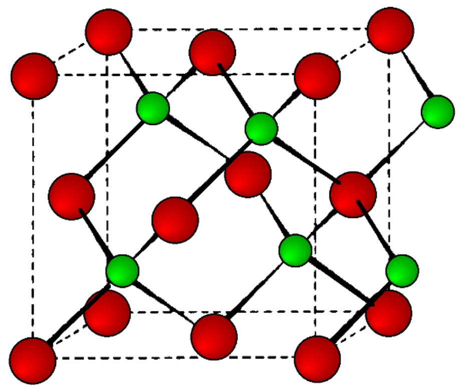
conduction Wannier function type

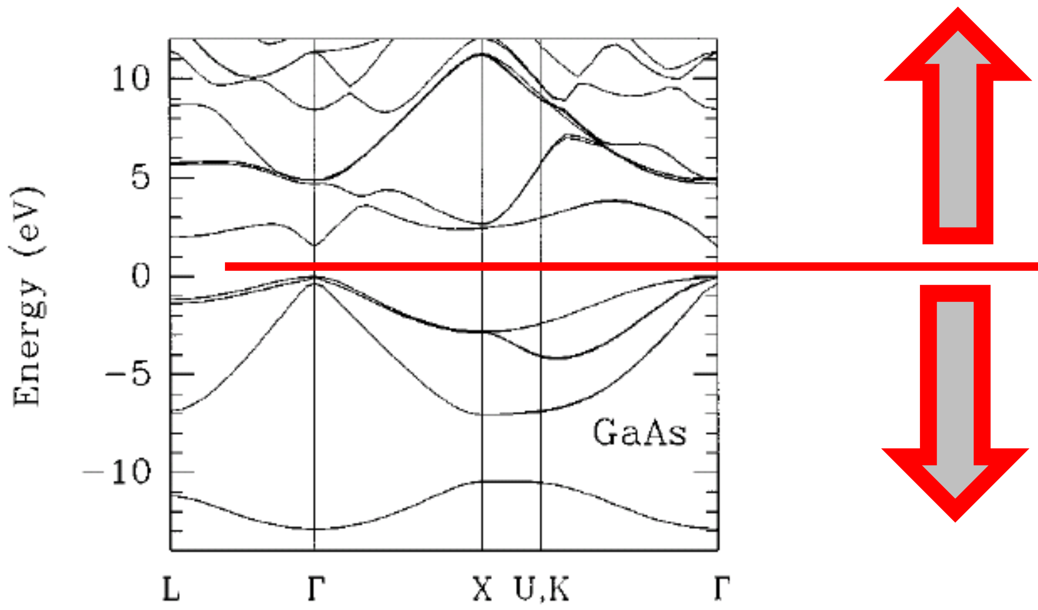
lattice site

unit cell volume

conduction bands

another unitary matrix





empty conduction bands

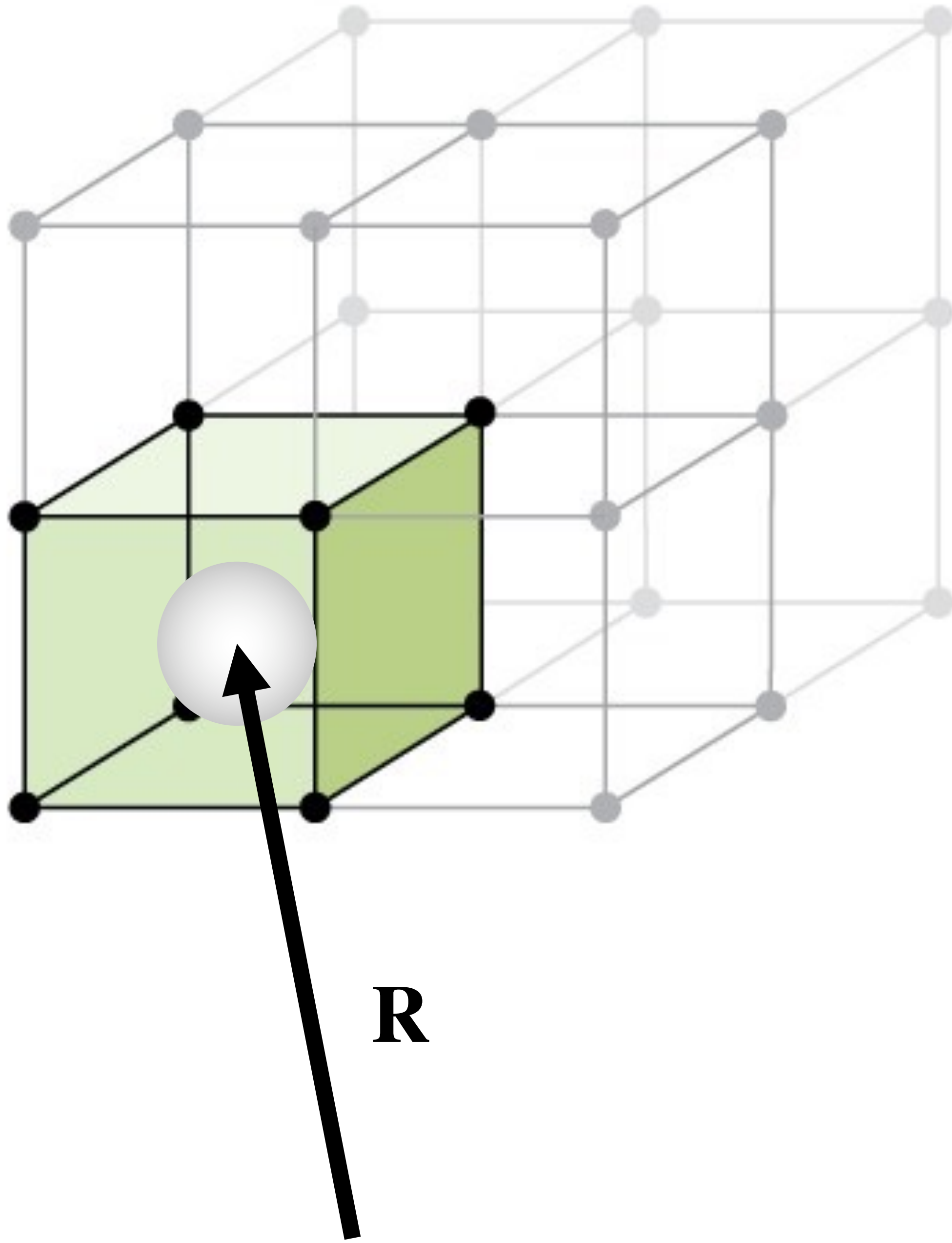
construct exponentially localized Wannier functions

filled valence bands

construct exponentially localized Wannier functions

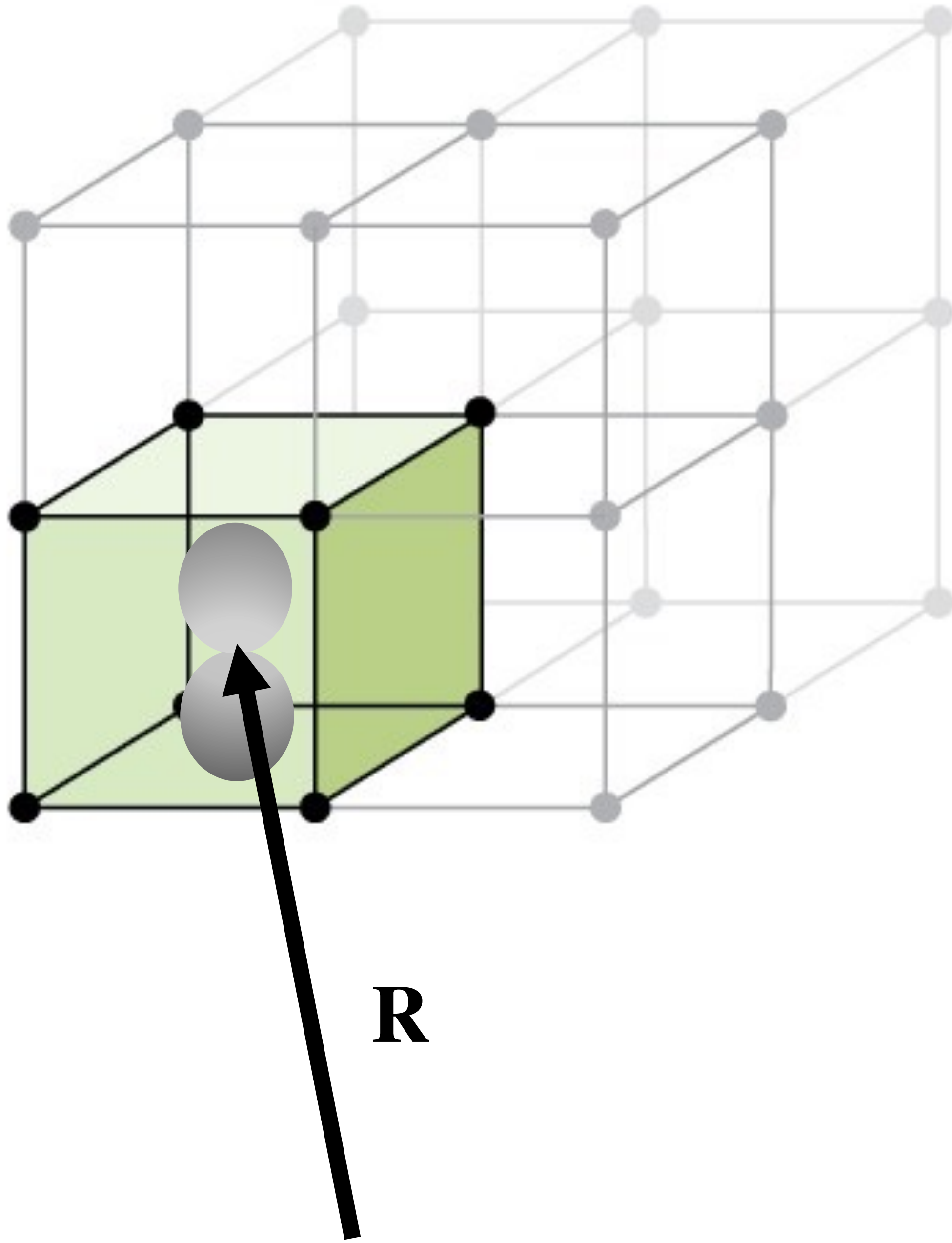
Bloch functions

$$\phi_{n\boldsymbol{k}}(\boldsymbol{x}) = \frac{u_{n\boldsymbol{k}}(\boldsymbol{x})}{(2\pi)^{3/2}} e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$$



Wannier function

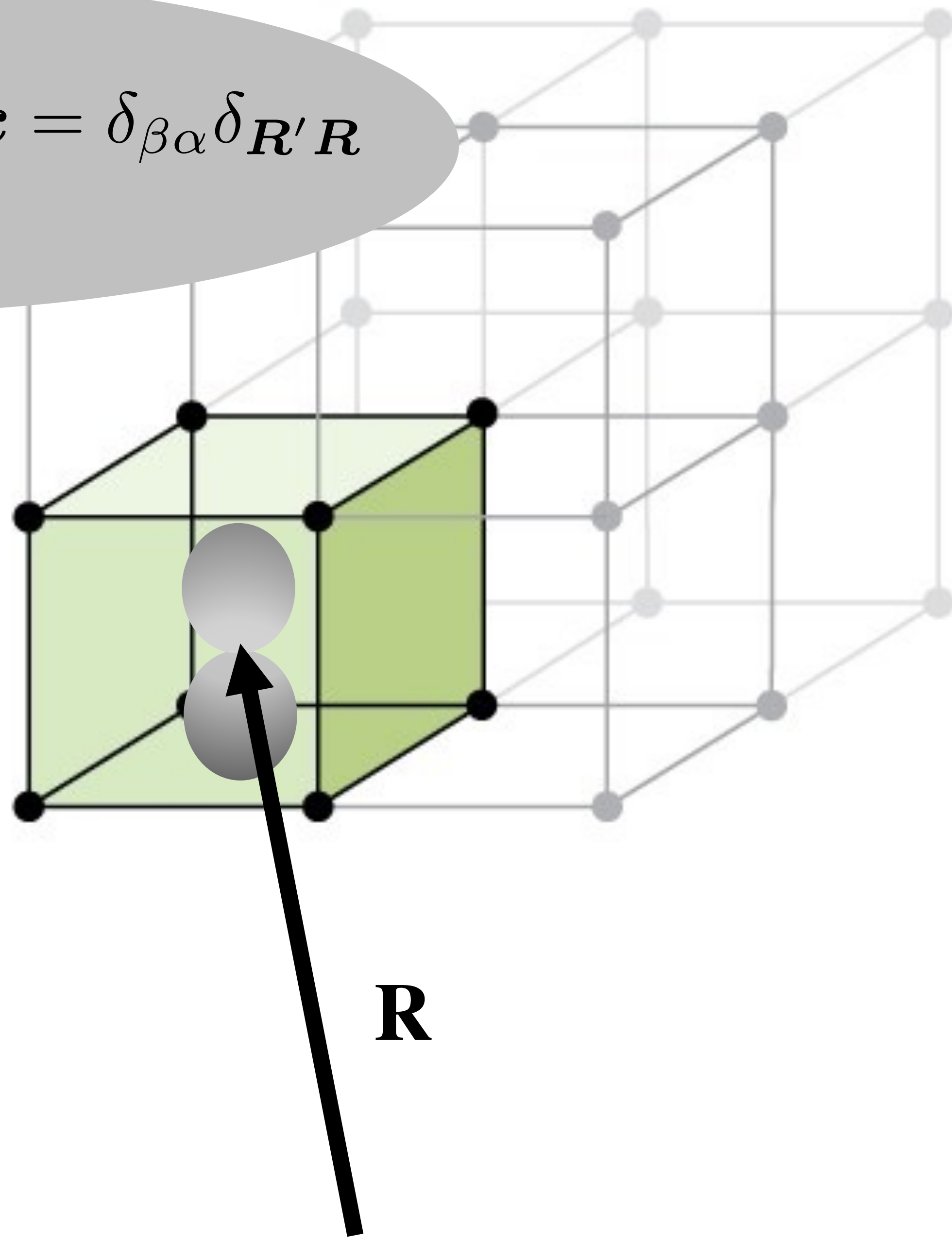
$$W_{1\mathbf{R}}(\mathbf{x})$$



Wannier function

$$W_{2\mathbf{R}}(\mathbf{x})$$

$$\int W^*_{\beta \boldsymbol{R}'}(\boldsymbol{x}) W_{\alpha \boldsymbol{R}}(\boldsymbol{x}) d\boldsymbol{x} = \delta_{\beta\alpha}\delta_{\boldsymbol{R}'\boldsymbol{R}}$$

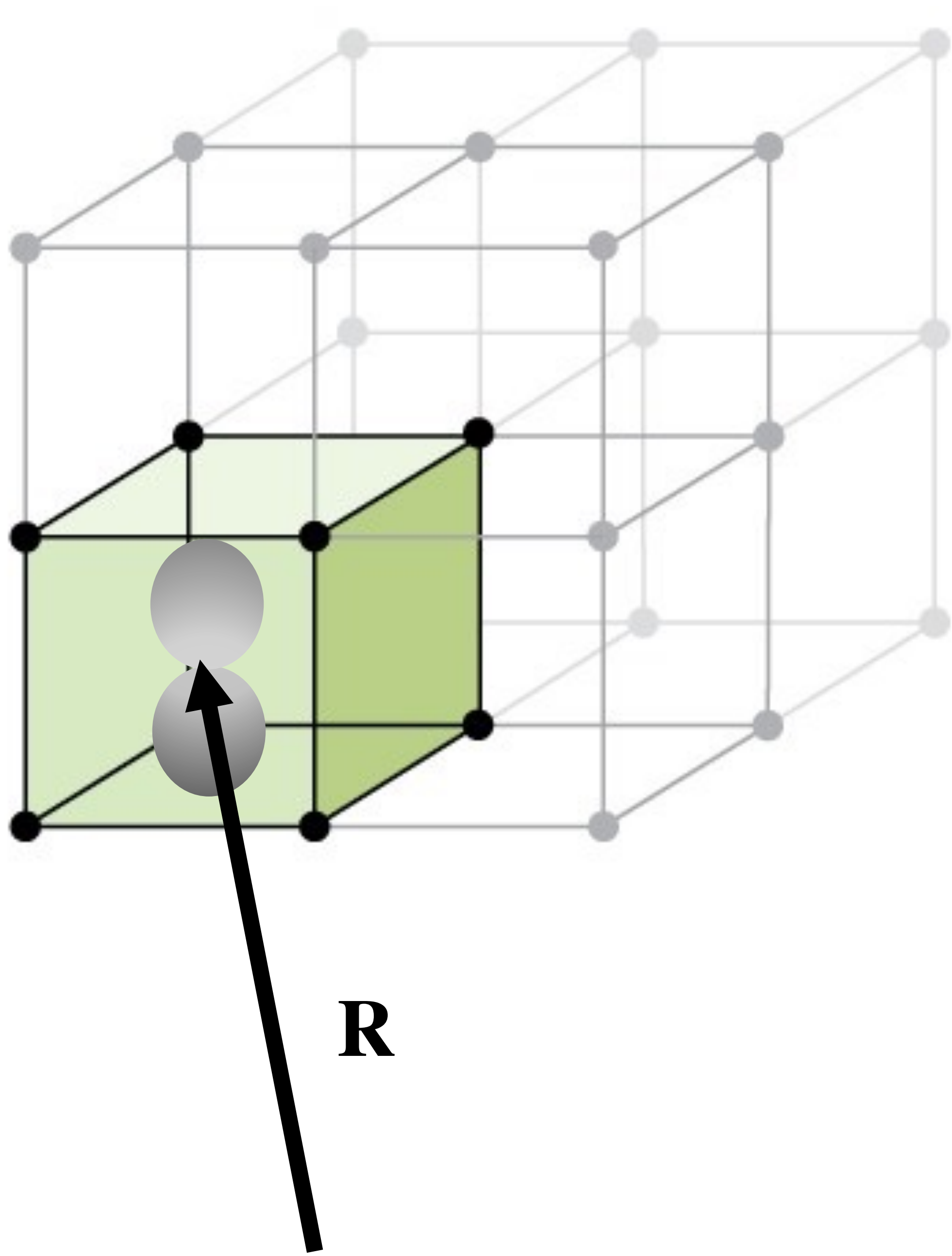


Wannier function

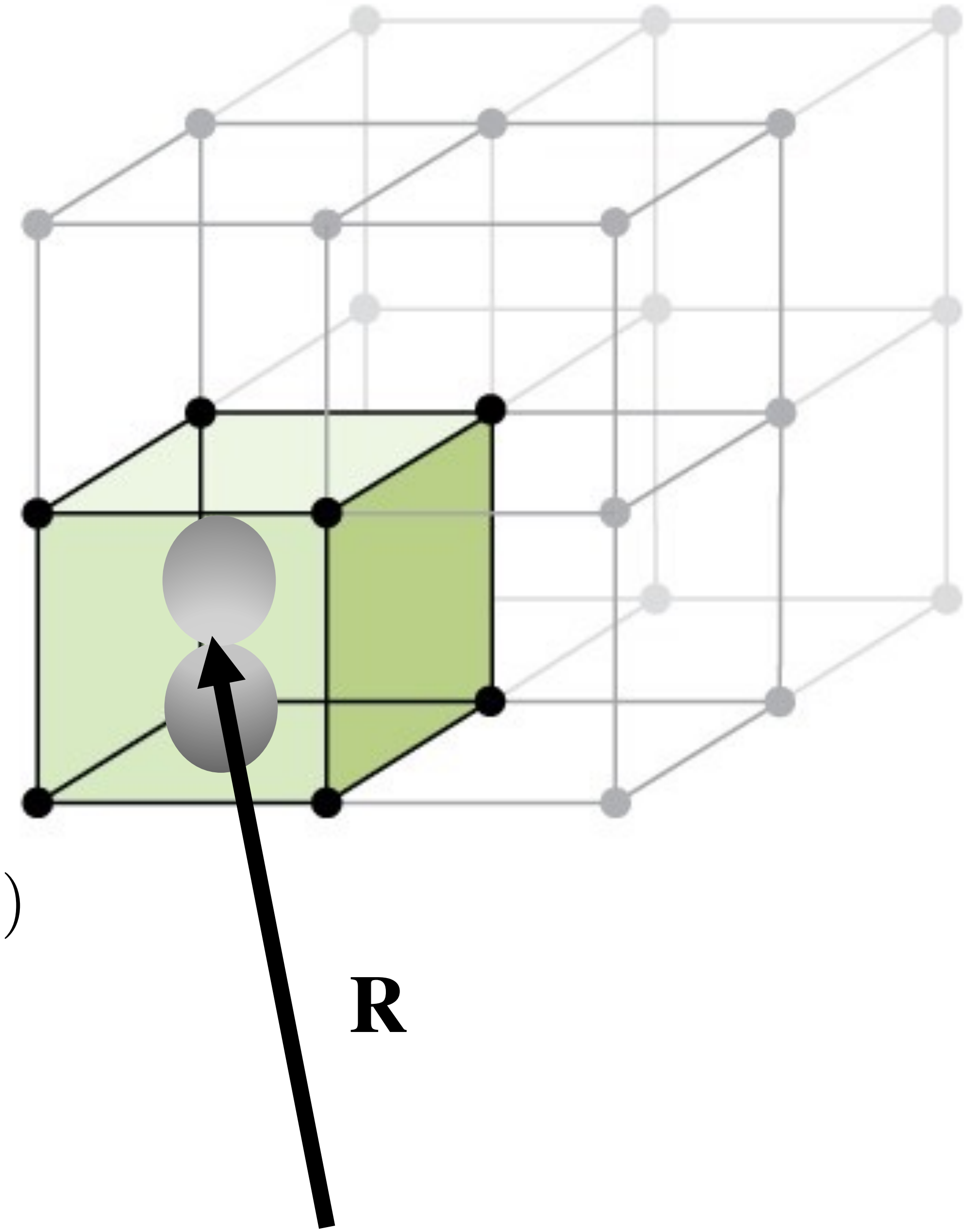
$$W_{2\mathbf{R}}(\mathbf{x})$$

$$\left\{ W_{\alpha \boldsymbol{R}}(\boldsymbol{x}) e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)} \right\}$$

not mutually orthogonal for the different R



adjust them
so they are

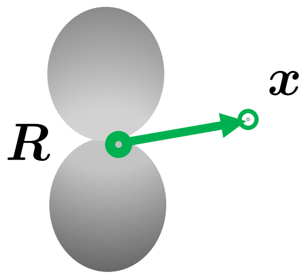


$$\psi(\boldsymbol{x},t) = \sum_{\alpha,\boldsymbol{R}} a_{\alpha\boldsymbol{R}}(t)\overline{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\left\{a_{\alpha\boldsymbol{R}}(t), a^{\dagger}_{\beta\boldsymbol{R}'}(t)\right\} = \delta_{\alpha\beta}\delta_{\boldsymbol{R}\boldsymbol{R}'}$$

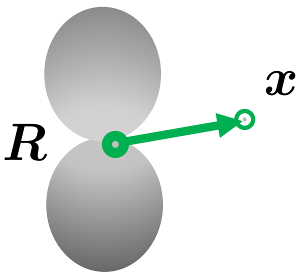
I. Mayer, Int. J. Quantum Chem. 90, 63 (2002)

$$\overline{W}_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t) = e^{i\Phi(\boldsymbol{x}, \boldsymbol{R}; t)} \chi_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t)$$



$$\Phi(\boldsymbol{x}, \boldsymbol{R}; t) = \frac{e}{\hbar c} \int s^i(\boldsymbol{w}; \boldsymbol{x}, \boldsymbol{R}) A^i(\boldsymbol{w}, t) d\boldsymbol{w}$$

$$\overline{W}_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t) = e^{i\Phi(\boldsymbol{x}, \boldsymbol{R}; t)} \chi_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t)$$



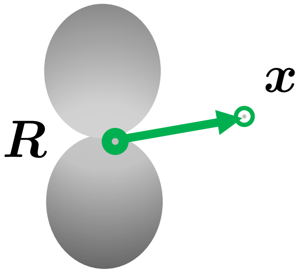
These functions are not mutually orthonormal, but are:

independent of $\mathbf{e}(\boldsymbol{y}, t)$

dependent only on $\mathbf{b}(\boldsymbol{y}, t)$

$$\Phi(\boldsymbol{x}, \boldsymbol{R}; t) = \frac{e}{\hbar c} \int s^i(\boldsymbol{w}; \boldsymbol{x}, \boldsymbol{R}) A^i(\boldsymbol{w}, t) d\boldsymbol{w}$$

$$\overline{W}_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t) = e^{i\Phi(\boldsymbol{x}, \boldsymbol{R}; t)} \chi_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t)$$



These functions are not mutually orthonormal, but are:

independent of $\boldsymbol{e}(\boldsymbol{y}, t)$

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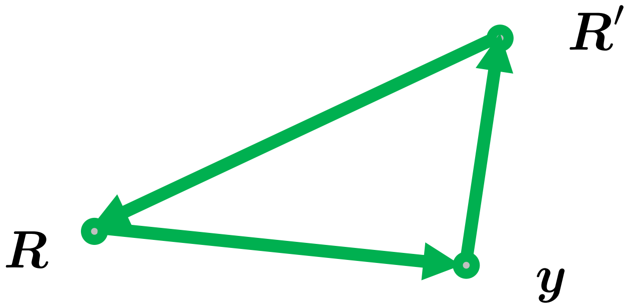
$$\Phi(\boldsymbol{x}, \boldsymbol{R}; t) = \frac{e}{\hbar c} \int s^i(\boldsymbol{w}; \boldsymbol{x}, \boldsymbol{R}) A^i(\boldsymbol{w}, t) d\boldsymbol{w}$$

$$\chi_{\alpha \boldsymbol{R}}(\boldsymbol{x}, t) = W_{\alpha \boldsymbol{R}}(\boldsymbol{x}) - \frac{1}{2} i \sum_{\beta, \boldsymbol{R}'} W_{\beta \boldsymbol{R}'}(\boldsymbol{x}) \int W^*_{\beta \boldsymbol{R}'}(\boldsymbol{y}) \Delta(\boldsymbol{R}', \boldsymbol{y}, \boldsymbol{R}; t) W_{\alpha \boldsymbol{R}}(\boldsymbol{y}) d\boldsymbol{y} + \cdots$$



$$\Delta(\boldsymbol{R}', \boldsymbol{y}, \boldsymbol{R}; t) = \Phi(\boldsymbol{R}, \boldsymbol{R}'; t) + \Phi(\boldsymbol{R}', \boldsymbol{y}; t) + \Phi(\boldsymbol{y}, \boldsymbol{R}; t)$$

Independent particle approximation

...a sketch of the guts of the calculation

field operator expansion

$$\psi(\boldsymbol{x},t) = \sum_{\alpha,\boldsymbol{R}} a_{\alpha\boldsymbol{R}}(t)\overline{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$$

effective density operator

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R'}}(t) = e^{i\Phi(\boldsymbol{R'},\boldsymbol{R};t)}\left\langle a^{\dagger}_{\beta\boldsymbol{R'}}(t)a_{\alpha\boldsymbol{R}}(t)\right\rangle$$

Independent particle approximation

…a sketch of the guts of the calculation

field operator expansion

$$\psi(\boldsymbol{x},t) = \sum_{\alpha,\boldsymbol{R}} a_{\alpha\boldsymbol{R}}(t)\overline{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$$

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dynamical equations

$$i\hbar\frac{\partial\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t} = \sum_{\lambda,\boldsymbol{R}''} e^{i\Delta(\boldsymbol{R},\boldsymbol{R}'',\boldsymbol{R}';t)}\left(H_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\eta_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t) - \eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)H_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)\right)$$
$$- e\Omega^{0}_{\boldsymbol{R}'}(\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'(t)}$$

Independent particle approximation

...a sketch of the guts of the calculation

field operator expansion

$$\psi(\boldsymbol{x},t) = \sum_{\alpha,\boldsymbol{R}} a_{\alpha\boldsymbol{R}}(t)\overline{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$$

effective density operator

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t) = e^{i\Phi(\boldsymbol{R}',\boldsymbol{R};t)}\left\langle a^{\dagger}_{\beta\boldsymbol{R}'}(t)a_{\alpha\boldsymbol{R}}(t)\right\rangle$$

dynamical equations

magnetic effects

magnetic and electric effects

$$i\hbar\frac{\partial\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t} = \sum_{\lambda,\boldsymbol{R}''} e^{i\Delta(\boldsymbol{R},\boldsymbol{R}'',\boldsymbol{R}';t)}\left(H_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\eta_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t) - \eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)H_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)\right)$$

$$- e\Omega^0_{\boldsymbol{R}'}(\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'(t)}$$

electric effects

Independent particle approximation

...a sketch of the guts of the calculation

field operator expansion

$$\psi(\boldsymbol{x},t) = \sum_{\alpha,\boldsymbol{R}} a_{\alpha\boldsymbol{R}}(t)\overline{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$$

effective density operator

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t) = e^{i\Phi(\boldsymbol{R}',\boldsymbol{R};t)}\left\langle a^{\dagger}_{\beta\boldsymbol{R}'}(t)a_{\alpha\boldsymbol{R}}(t)\right\rangle$$

dynamical equations

magnetic effects

magnetic and electric effects

$$i\hbar\frac{\partial\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t} = \sum_{\lambda,\boldsymbol{R}''} e^{i\Delta(\boldsymbol{R},\boldsymbol{R}'',\boldsymbol{R}';t)}\left(H_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\eta_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t) - \eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)H_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)\right)$$
$$- e\Omega^{0}_{\boldsymbol{R}'}(\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'(t)}$$

electric effects

only dependent on electromagnetic fields, not potentials
valid for arbitrary $\mathrm{e}(\boldsymbol{y},t)$ ***and*** $\mathrm{b}(\boldsymbol{y},t)$

Independent particle approximation

...a sketch of the guts of the calculation

Green function for site $\boldsymbol{R}$

$$G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t) = \frac{i}{2}\sum_{\alpha,\beta,\boldsymbol{R}'} \eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)\, e^{i\Delta(\boldsymbol{R}',\boldsymbol{y},\boldsymbol{R};t)}\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{y},t)\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$$

$$+\frac{i}{2}\sum_{\alpha,\beta,\boldsymbol{R}'} \eta_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}}(t)\, e^{i\Delta(\boldsymbol{R},\boldsymbol{x},\boldsymbol{R}';t)}\chi^*_{\alpha\boldsymbol{R}}(\boldsymbol{y},t)\chi_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)$$

Site charge and current densities

$$\rho_{\boldsymbol{R}}(\boldsymbol{x},t) = -ie\left[G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)\right]_{\boldsymbol{y}\to\boldsymbol{x}}$$

$$\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t) = \left[\mathcal{J}_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)\right]_{\boldsymbol{y}\to\boldsymbol{x}}$$

with

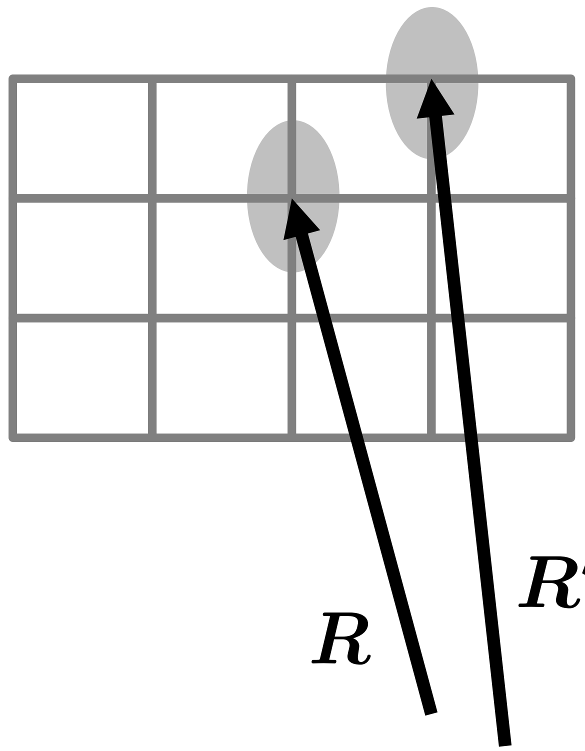
$$\langle\rho(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}}\rho_{\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\langle\boldsymbol{j}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}}\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

site charge and current densities

$$\langle \rho(\boldsymbol{x}, t) \rangle = \sum_{\boldsymbol{R}} \rho_{\boldsymbol{R}}(\boldsymbol{x}, t)$$

$$\langle \boldsymbol{j}(\boldsymbol{x}, t) \rangle = \sum_{\boldsymbol{R}} \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x}, t)$$



site charge and current densities

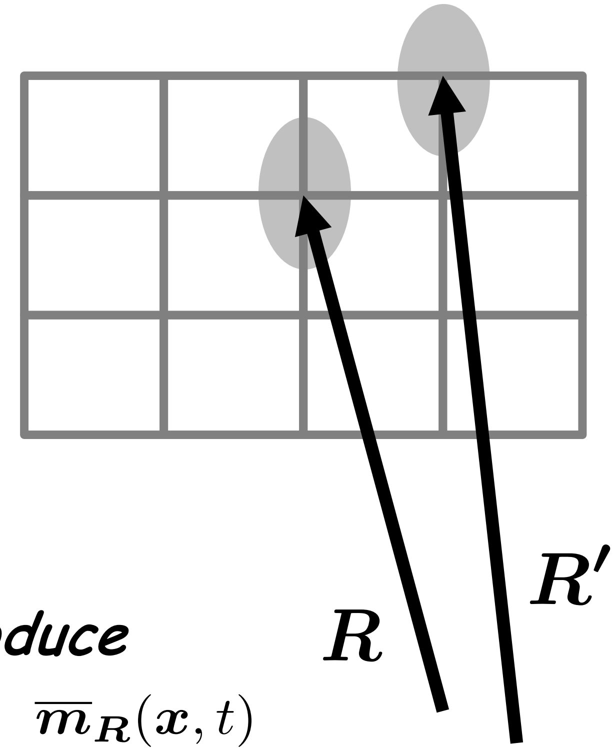
$$\langle \rho(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \rho_{\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\langle \boldsymbol{j}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

introduce

$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ $\quad$ $\overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$

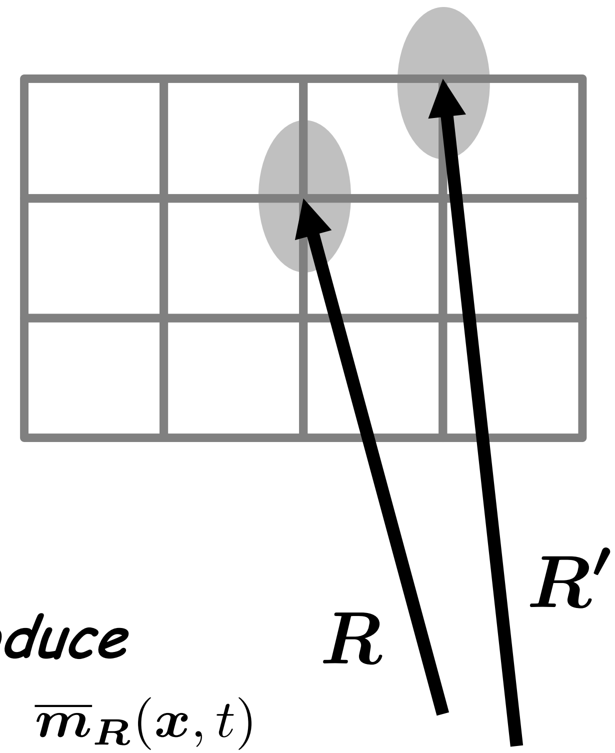
as for an atom



site charge and current densities

$$\langle\rho(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \rho_{\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\langle\boldsymbol{j}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$$



introduce

$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ $\qquad$ $\overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$

as for an atom

but

$$\frac{\partial\rho_{\boldsymbol{R}}(\boldsymbol{x},t)}{\partial t} + \nabla\cdot\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t) \neq 0$$

site charge and current densities

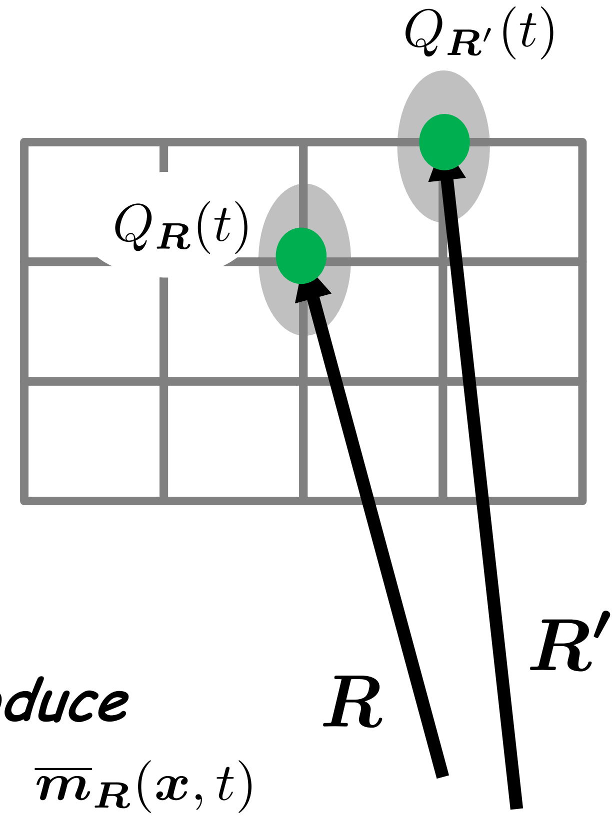
$$\langle \rho(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \rho_{\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\langle \boldsymbol{j}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

introduce

$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ $\overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$

as for an atom



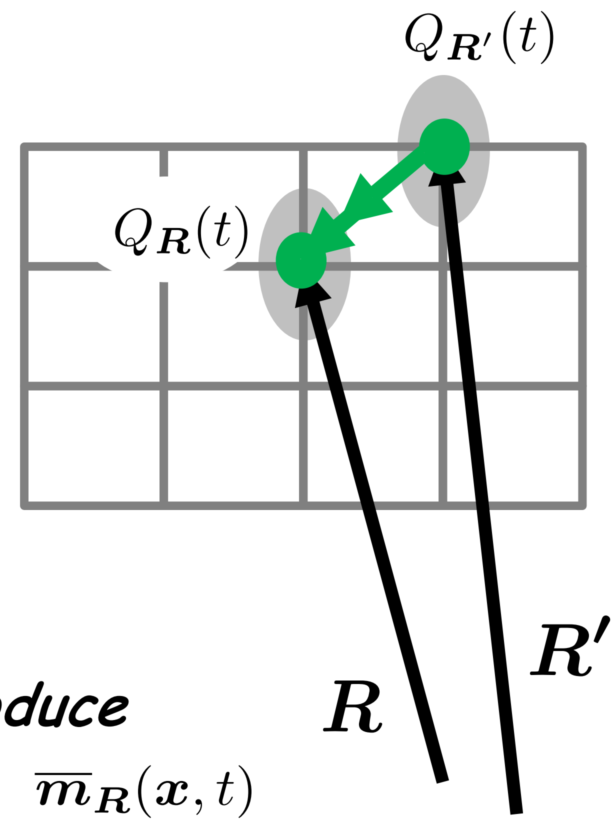
introduce a "free" charge density

$$\rho_{free}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}} Q_{\boldsymbol{R}}(t)\delta(\boldsymbol{x}-\boldsymbol{R})$$

site charge and current densities

$$\langle \rho(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \rho_{\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\langle \boldsymbol{j}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$$



introduce

$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ $\quad \overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$

as for an atom

introduce a "free" charge density

$$\rho_{free}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}} Q_{\boldsymbol{R}}(t)\delta(\boldsymbol{x}-\boldsymbol{R})$$

$$\frac{dQ_{\boldsymbol{R}}(t)}{dt} = \sum_{\boldsymbol{R}'} I(\boldsymbol{R},\boldsymbol{R}';t)$$

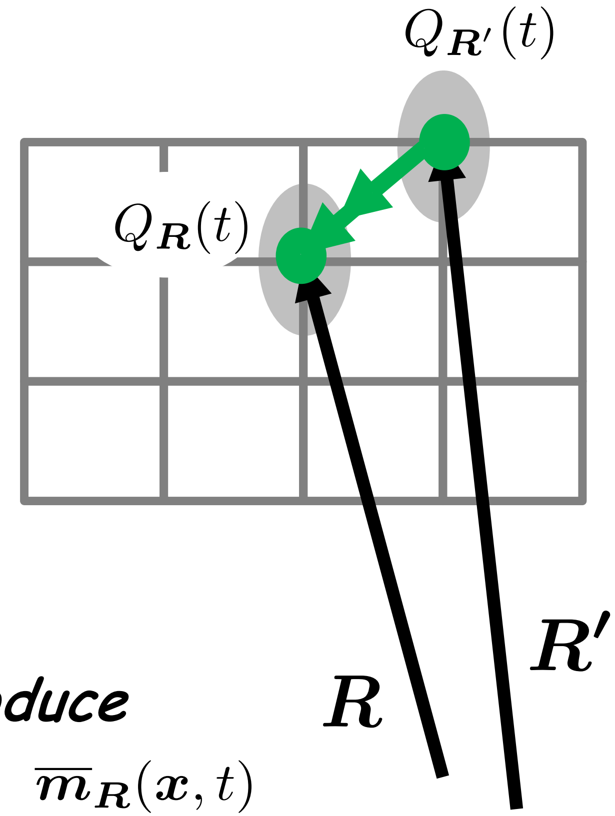
and a link current density

$$\boldsymbol{j}_{free}(\boldsymbol{x},t) = \frac{1}{2}\sum_{\boldsymbol{R},\boldsymbol{R}'} \boldsymbol{s}(\boldsymbol{x};\boldsymbol{R},\boldsymbol{R}')I(\boldsymbol{R},\boldsymbol{R}';t)$$

site charge and current densities

$$\langle \rho(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \rho_{\boldsymbol{R}}(\boldsymbol{x},t)$$

$$\langle \boldsymbol{j}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}} \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$$



introduce

$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ $\quad \overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$

as for an atom

introduce a "free" charge density

$$\rho_{free}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}} Q_{\boldsymbol{R}}(t)\delta(\boldsymbol{x}-\boldsymbol{R})$$

and a link current density

$$\boldsymbol{j}_{free}(\boldsymbol{x},t)$$

and a new contribution

$$\widetilde{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

to each site magnetization

$$\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t) = \overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t) + \widetilde{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

microscopic fields

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{j}(\boldsymbol{x},t) = \frac{\partial \boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla \times \boldsymbol{m}(\boldsymbol{x},t) + \boldsymbol{j}_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{p}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}} \boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

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microscopic fields

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Vanish for a trivial insulator in linear response

$$\boldsymbol{p}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}} \boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

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microscopic fields

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$$\boldsymbol{m}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}} \boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

For the ground state, the integrals of these divided by the volume of the unit cell yield

$$\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t) = \overline{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t) + \widetilde{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

The "atomic" magnetization of the modern theory

The "itinerant" magnetization of the modern theory

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

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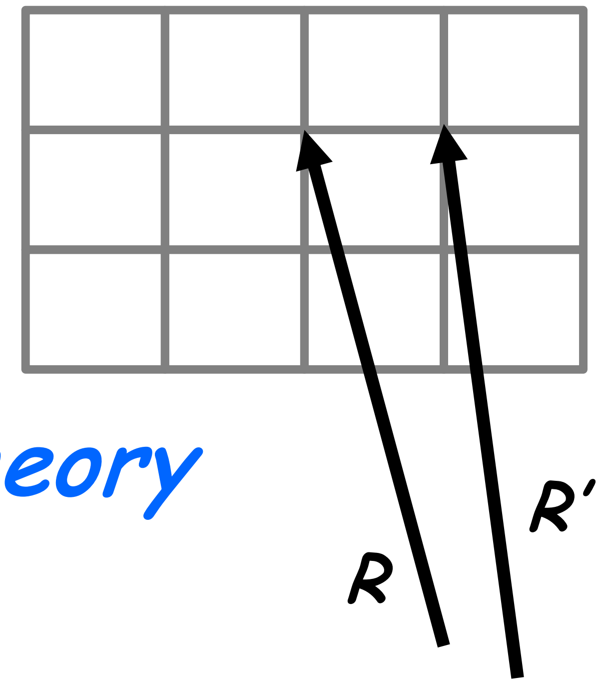

Generalized lattice gauge theory

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

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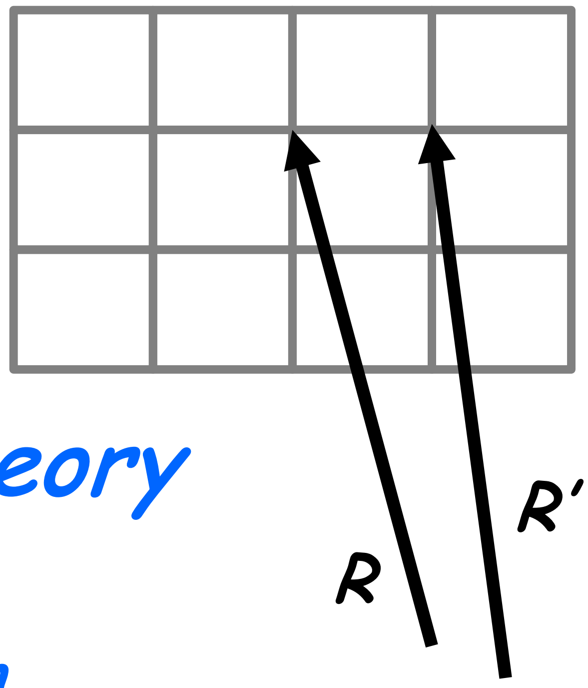

Generalized lattice gauge theory

→ *arbitrary variation of vector and scalar potentials between sites*

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

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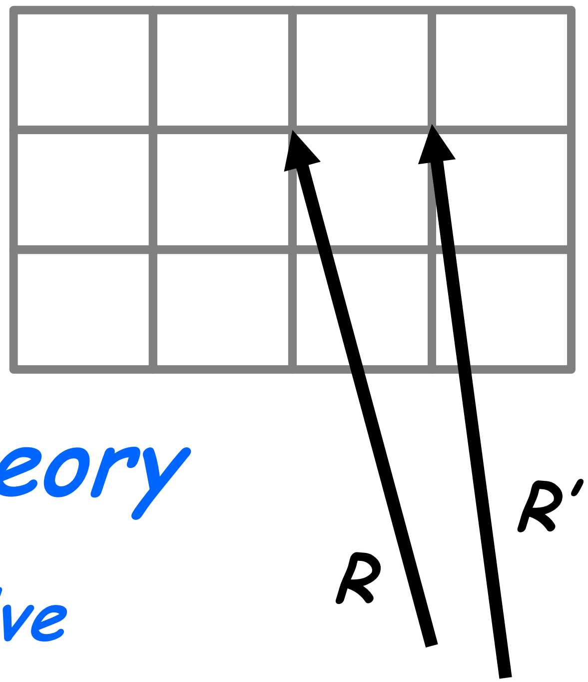

Generalized lattice gauge theory

→ *hopping matrix elements involve a set of states at each site*

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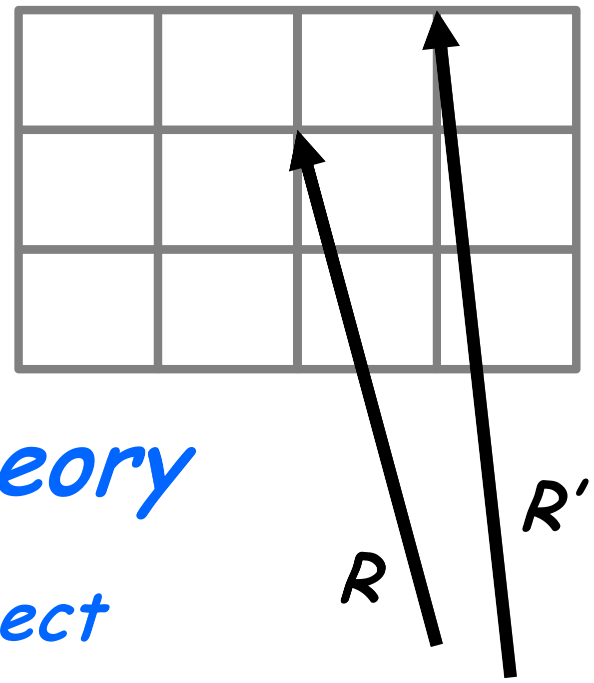

Generalized lattice gauge theory

→ *hopping matrix elements connect each site to (in principle) every other site*

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

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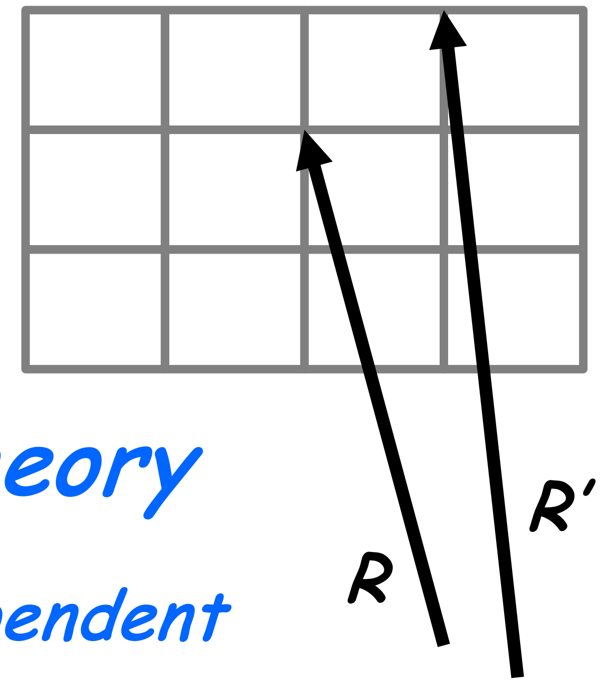


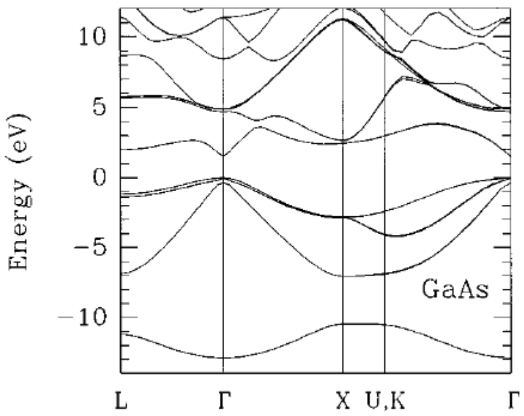
Generalized lattice gauge theory

→ *matrix elements are time dependent but gauge independent; involves dynamics at each site as well as between*

Overview
The story for molecules and atoms
Generalizing to condensed matter
Some results
Perspective

Ground states

topologically trivial insulators



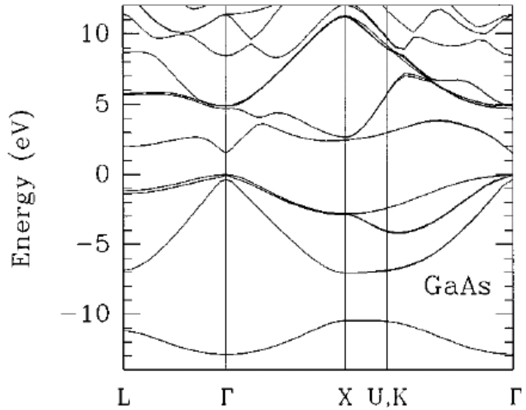
P and M agree with those of the "modern theory"

Phys. Rev. B99, 235140 (2019)
Phys. Rev. Res. 2, 033126 (2020)

metals

Ground states

topologically trivial insulators



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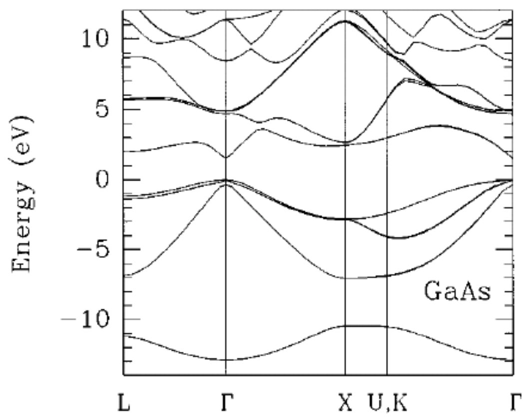

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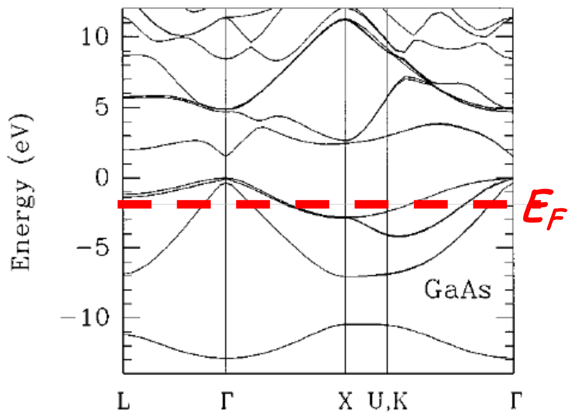

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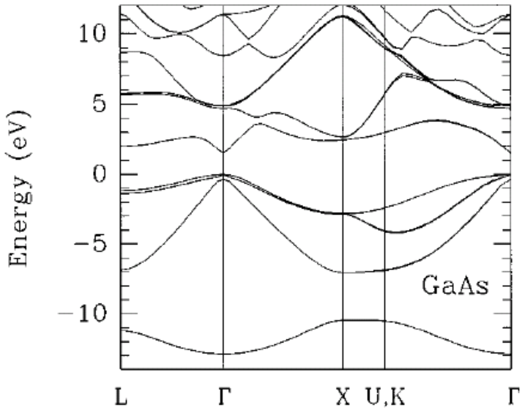

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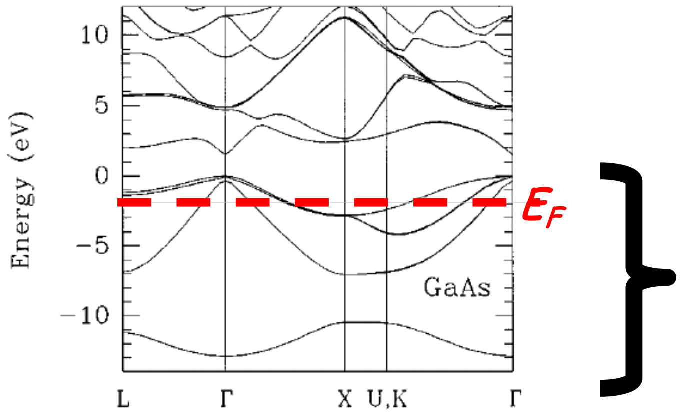
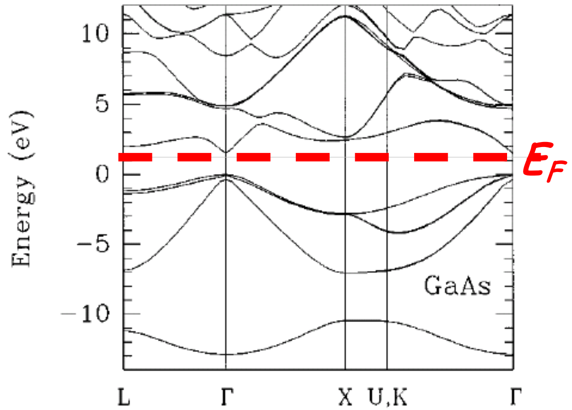


use full set of valence bands to form Wannier functions

SciPost Phys. 14, 058 (2023)

Ground states

topologically trivial insulators



***P* and *M* agree with those of the "modern theory"**

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$$\eta_{\alpha \boldsymbol{R};\beta \boldsymbol{R}'} = f_\alpha \delta_{\alpha\beta} \delta_{\boldsymbol{R}\boldsymbol{R}'}$$

$$f_\alpha = 0 \text{ or } 1$$

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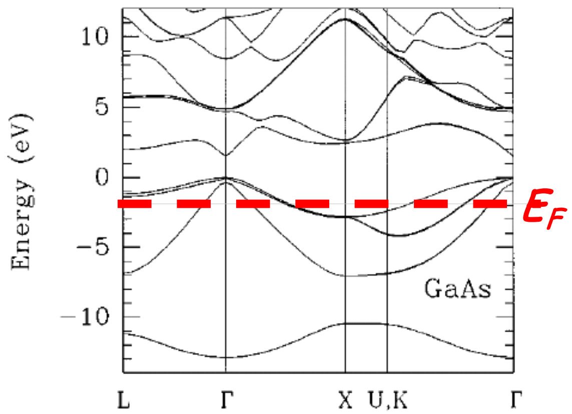

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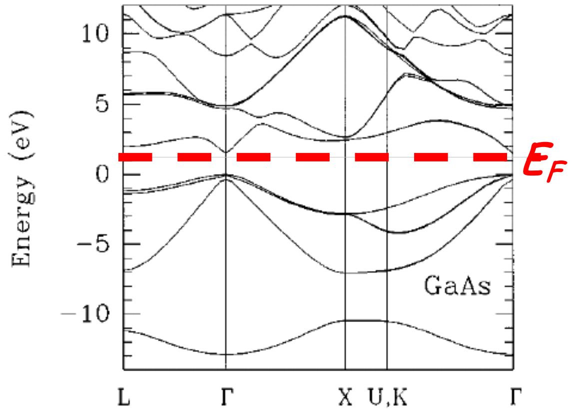

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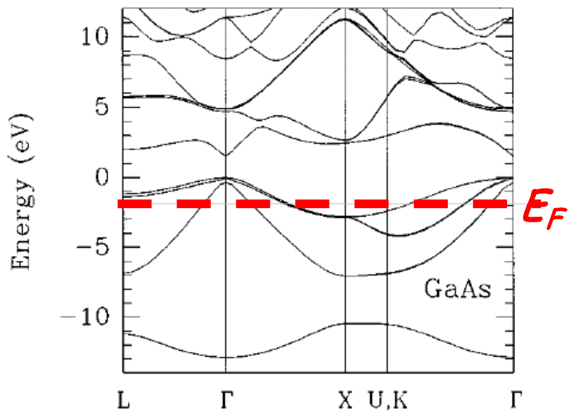

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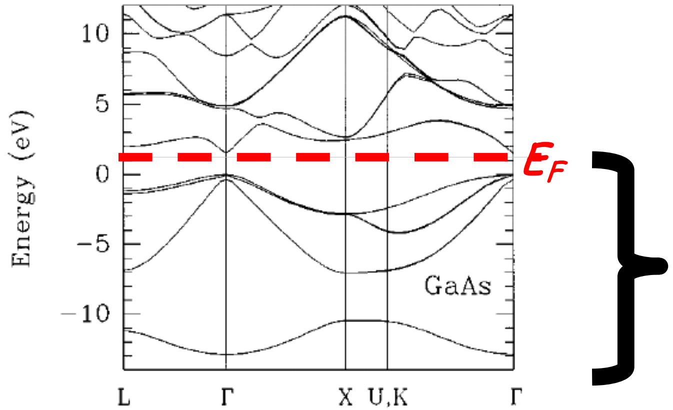

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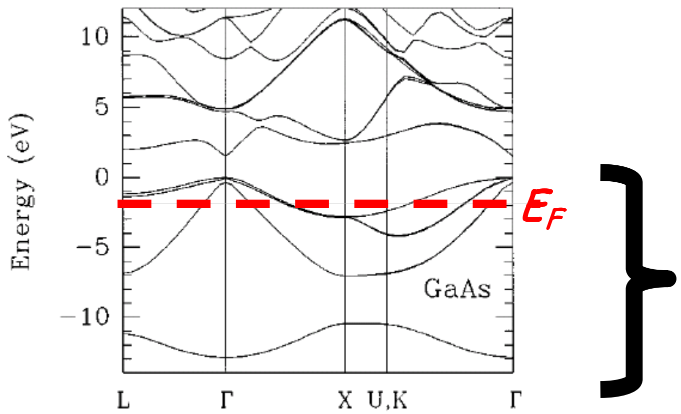

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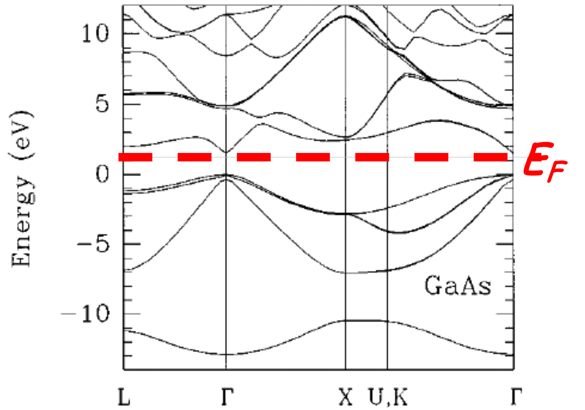
calculate results in Wannier basis, then convert to Bloch state basis

Ground states

topologically trivial insulators



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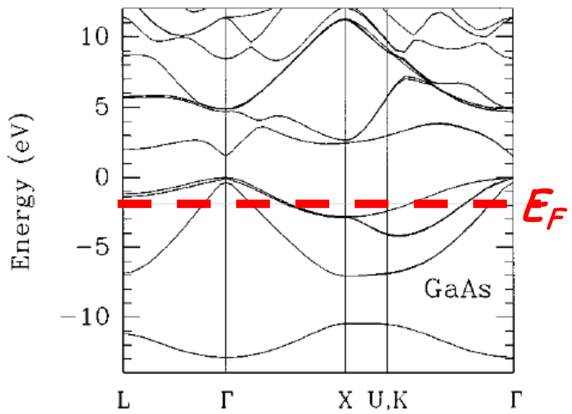

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general
approach

calculate results in Wannier basis,
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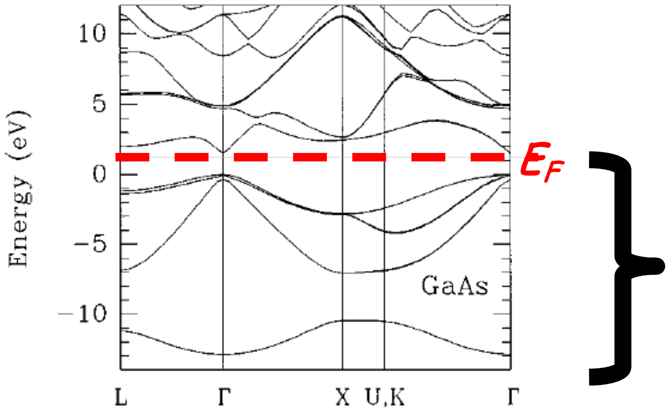
Ground states

topologically trivial insulators



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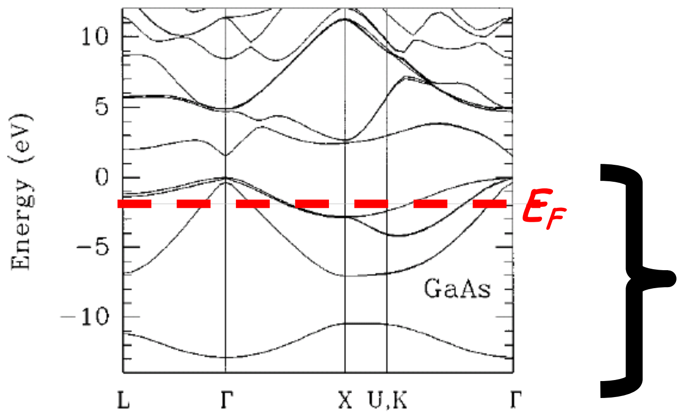

use full set of valence bands to form Wannier functions

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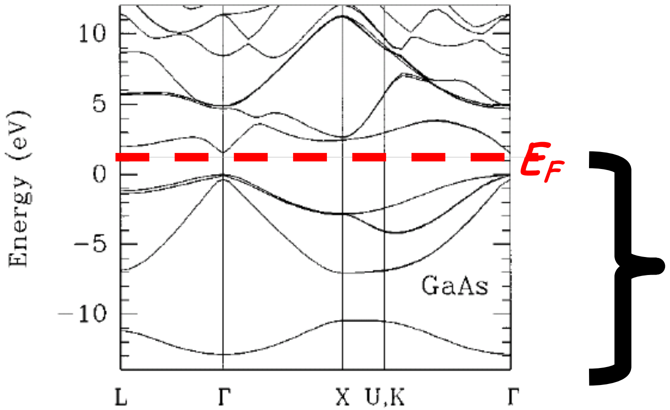
$$\xi^i_{nm}(\boldsymbol{k}) = \frac{i}{\Omega_{uc}} \int_{uc} u^*_{n\boldsymbol{k}}(\boldsymbol{x}) \frac{\partial u_{m\boldsymbol{k}}(\boldsymbol{x})}{\partial k^i} d\boldsymbol{x}$$

$$\mathcal{W}^i_{nm}(\boldsymbol{k}) = i \sum_{\alpha} \left(\partial_i U_{n\alpha}(\boldsymbol{k})\right) U^\dagger_{\alpha m}(\boldsymbol{k})$$

Ground states

topologically trivial insulators



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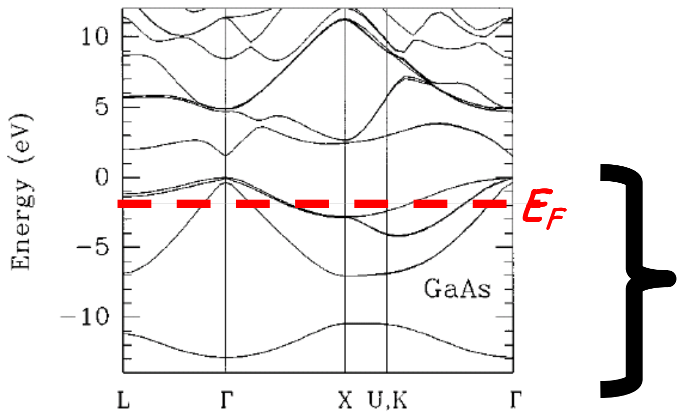
$$P^i = e\int \frac{d\boldsymbol{k}}{(2\pi)^3}\sum_n f_n\left(\xi^i_{nn}(\boldsymbol{k}) + \mathcal{W}^i_{nn}(\boldsymbol{k})\right)$$

+ ion contribution

metals

(well, p-doped semiconductors)



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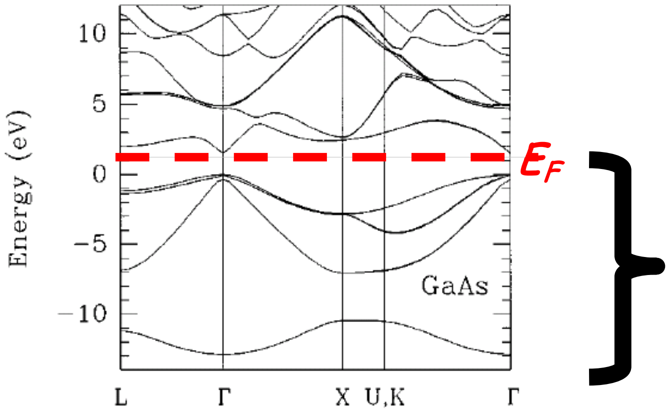
topologically trivial insulators



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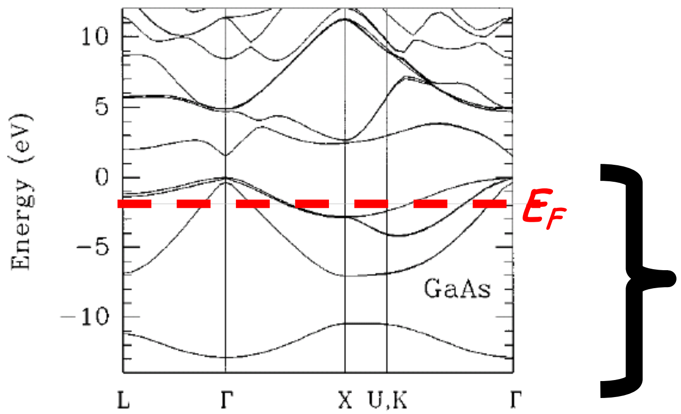

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Ground states

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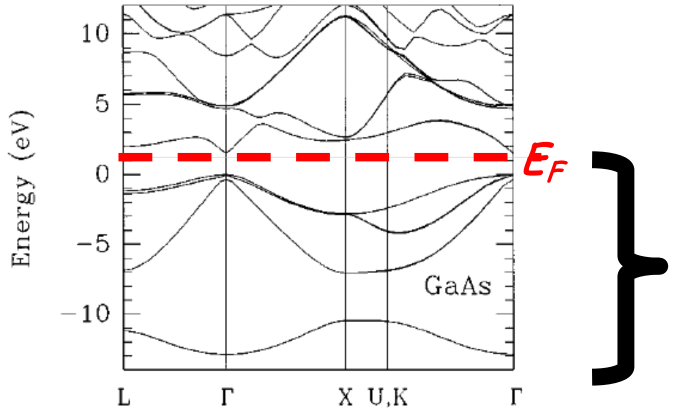
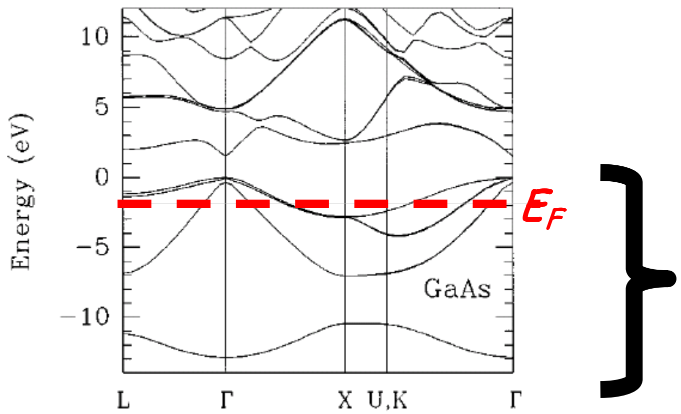
topologically trivial insulators





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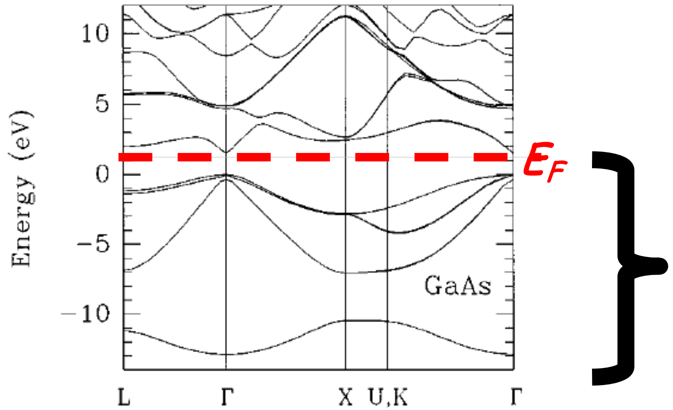
gauge dependence ⟺ *"quantum of ambiguity"*

Ground states

topologically trivial insulators



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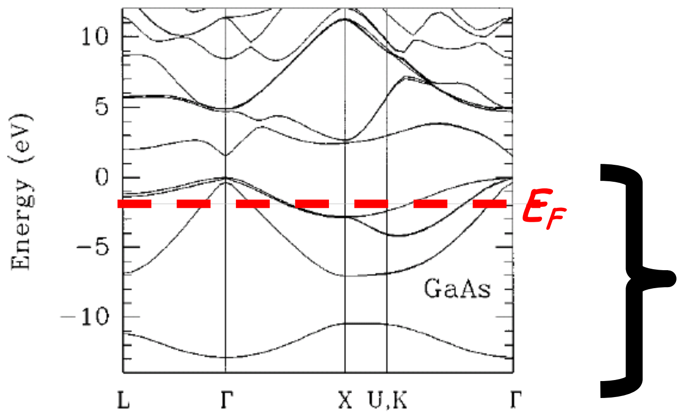

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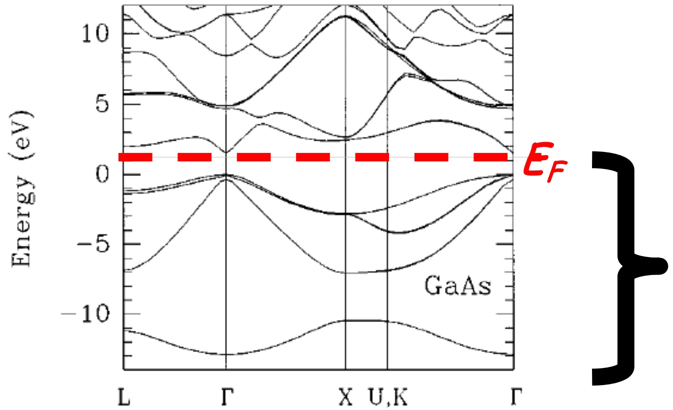
presumably a more general gauge dependence

gauge dependence ⟺ "quantum of ambiguity"

Ground states

topologically trivial insulators



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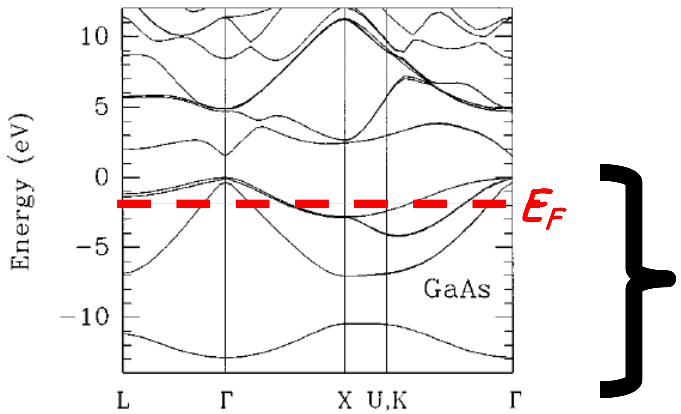

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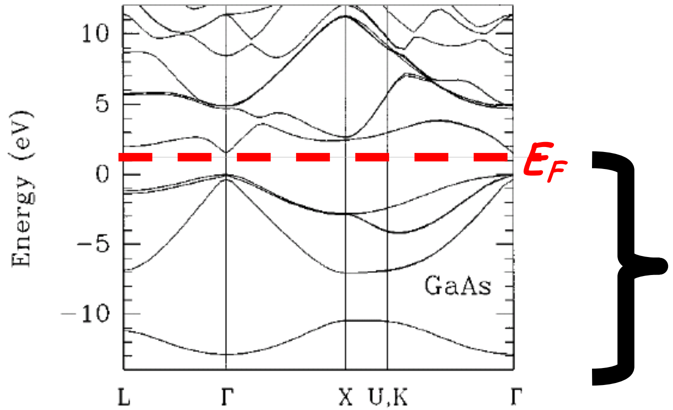
as opposed to "modern theory,"
where P not defined:
Phys. Rev. Lett. 80, 1800 (1998)
Phys. Rev. Lett. 82, 370 (1999)

Ground states

topologically trivial insulators



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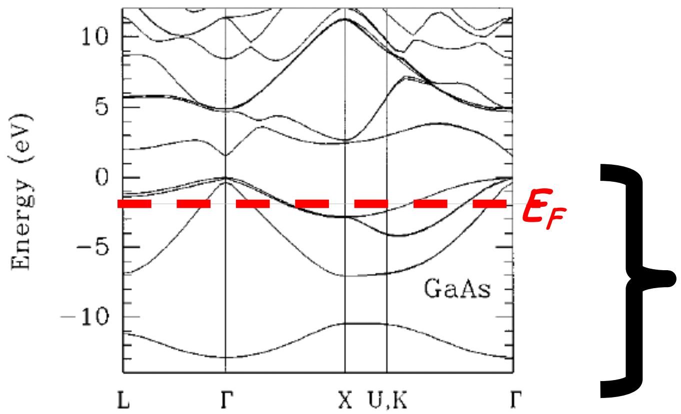

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SciPost Phys. 14, 058 (2023)

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Phys. Rev. Lett. 80, 1800 (1998)
Phys. Rev. Lett. 82, 370 (1999)

Our M and the "modern theory" M are different, both in approach to definition and in result

Phys. Rev. B74, 024408 (2006)
Phys. Rev. Lett. 99, 197202 (2007)

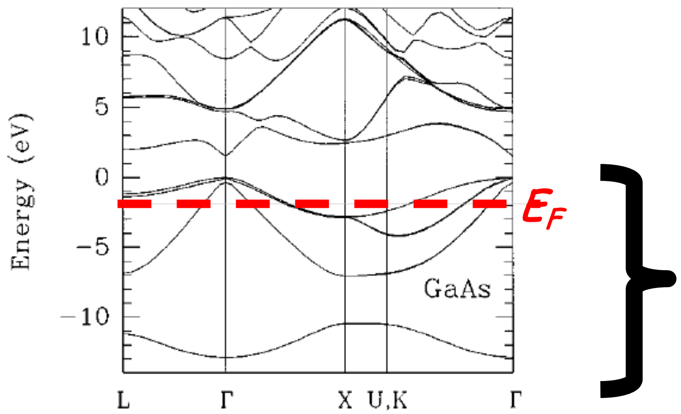
Ground states

Chern insulators

Chern invariant of valence bands nonzero

metals

(well, p-doped semiconductors)



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SciPost Phys. 14, 058 (2023)

$$P^i = e\int \frac{d\boldsymbol{k}}{(2\pi)^3}\sum_n f_{n\boldsymbol{k}}\left(\xi^i_{nn}(\boldsymbol{k}) + \mathcal{W}^i_{nn}(\boldsymbol{k})\right)$$

as opposed to "modern theory,"
where P not defined:

Phys. Rev. Lett. 80, 1800 (1998)
Phys. Rev. Lett. 82, 370 (1999)

Our M and the "modern theory"
M are different, both in approach
to definition and in result

Phys. Rev. B74, 024408 (2006)
Phys. Rev. Lett. 99, 197202 (2007)

Ground states

Chern insulators

Chern invariant of valence bands nonzero

→

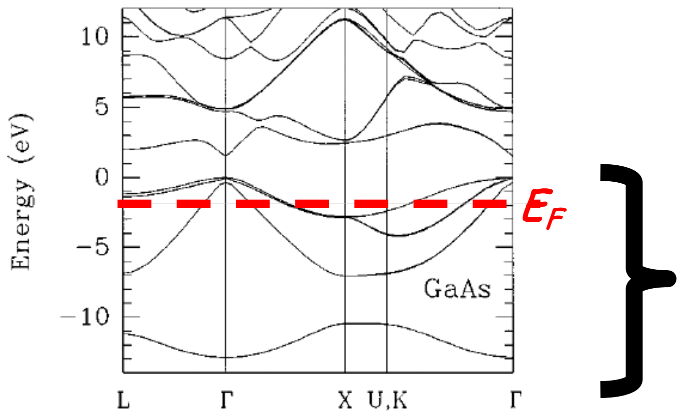
make Wannier localized Wannier functions out of an expanded set of bands where Chern invariant does vanish

$$P^i = e \int \frac{d\boldsymbol{k}}{(2\pi)^3} \sum_n f_n \left(\xi^i_{nn}(\boldsymbol{k}) + \mathcal{W}^i_{nn}(\boldsymbol{k}) \right)$$

Phys. Rev. B107, 115110 (2023)

metals

(well, p-doped semiconductors)



use full set of valence bands to form Wannier functions

SciPost Phys. 14, 058 (2023)

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Phys. Rev. B107, 115110 (2023)

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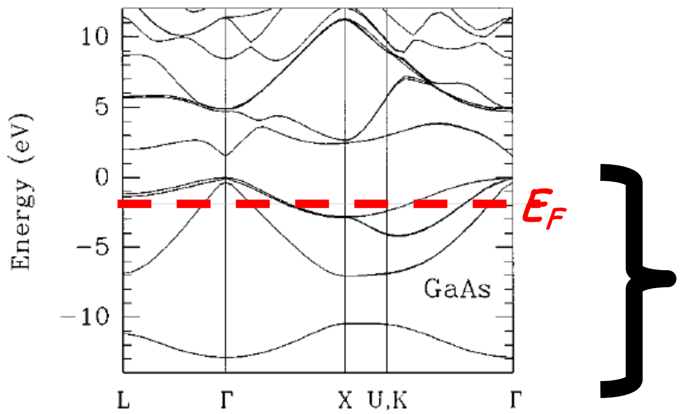

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SciPost Phys. 14, 058 (2023)

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Chern invariant of valence bands nonzero

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Phys. Rev. B107, 115110 (2023)

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>
> *Phys. Rev. B74, 024408 (2006)*
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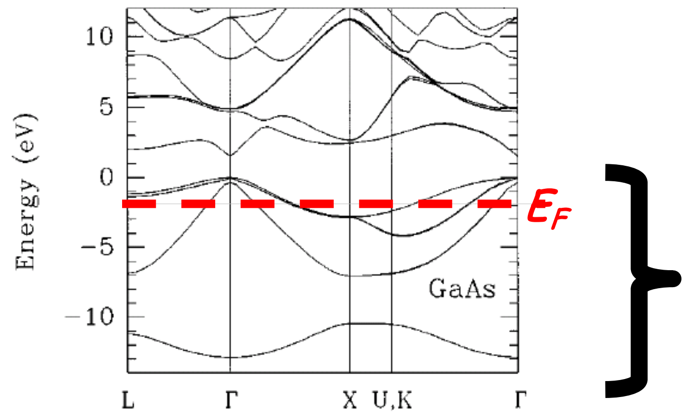
both results are insensitive to

$$E_{m\boldsymbol{k}} \to E_{m\boldsymbol{k}} + \epsilon$$

metals

(well, p-doped semiconductors)



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SciPost Phys. 14, 058 (2023)

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Linear response

"Long wavelength" limit:

$$\boldsymbol{E}(\boldsymbol{x},t) \rightarrow \boldsymbol{E}(t)$$

$$\boldsymbol{B}(\boldsymbol{x},t) \rightarrow 0$$

general approach

calculate results in Wannier basis, then convert to Bloch state basis

Linear response

"Long wavelength" limit:

$$\boldsymbol{E}(\boldsymbol{x},t) \to \boldsymbol{E}(t)$$

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$$\boldsymbol{J}(t) = \boldsymbol{J}_{free}(t) + \frac{d\boldsymbol{P}(t)}{dt}$$

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in general both will contribute

$$J^i(\omega) = \sigma^{ij}(\omega) E^j(\omega)$$

In general:

gauge dependent

$$\boldsymbol{J}(\omega) = \boldsymbol{J}_{free}(\omega) - i\omega \boldsymbol{P}(\omega)$$

gauge independent

general approach

calculate results in Wannier basis, then convert to Bloch state basis

Linear response

$$\boldsymbol{E}(\boldsymbol{x},t) \to \boldsymbol{E}(t)$$

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in general both will contribute

$$J^i(\omega) = \sigma^{ij}(\omega)E^j(\omega)$$

Insulator

$$\sigma^{ij}(\omega) = -i\omega\frac{e^2}{\hbar}\sum_{n,m}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\frac{f_m\omega_{nm}(\xi^i_{mn}\xi^j_{nm} + \xi^j_{mn}\xi^i_{nm})}{(\omega^2_{nm} - \omega^2)}$$

$$- i\frac{e^2}{\hbar}\sum_{n,m}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\frac{f_m\omega^2_{nm}(\xi^i_{mn}\xi^j_{nm} - \xi^j_{mn}\xi^i_{nm})}{(\omega^2_{nm} - \omega^2)}$$

Phys. Rev. B99, 235140 (2019)

Linear response

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$$\lim_{\omega\rightarrow 0} \sigma^{ij}(\omega) = \frac{e^2}{\hbar} \sum_{m} \int \frac{d\boldsymbol{k}}{(2\pi)^3} f_m \left(\frac{\partial \xi^i_{mm}}{\partial k^j} - \frac{\partial \xi^j_{mm}}{\partial k^i} \right)$$

quantized anomalous Hall current

Phys. Rev. B99, 235140 (2019)

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quantized anomalous Hall current

from "free current"

Phys. Rev. B99, 235140 (2019)

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$$\boldsymbol{J}(t) = \boldsymbol{J}_{free}(t) + \frac{d\boldsymbol{P}(t)}{dt}$$

in general both will contribute

$$J^i(\omega) = \sigma^{ij}(\omega)E^j(\omega)$$

Topologically trivial insulator

$$\boldsymbol{J}_{free}(t) = 0$$

$$\boldsymbol{P}(\omega) = \frac{e^2}{\hbar}\sum_{n,m}\int \frac{d\boldsymbol{k}}{(2\pi)^3}\frac{f_{mn}\boldsymbol{\xi}_{mn}(\boldsymbol{k})\left(\boldsymbol{\xi}_{nm}(\boldsymbol{k})\cdot\boldsymbol{E}(\omega)\right)}{(\omega_{nm}(\boldsymbol{k})-\omega)}$$

Phys. Rev. B99, 235140 (2019)

Linear response

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in general both will contribute

$$J^i(\omega) = \sigma^{ij}(\omega)E^j(\omega)$$

Metal

If time reversal symmetry:

$\boldsymbol{J}_{free}(\omega)$	$\boldsymbol{P}(\omega)$
diverges as $\omega \to 0$	finite as $\omega \to 0$
"intraband"	"interband"

SciPost Phys. 14, 058 (2023)

Linear response

$$\boldsymbol{E}(\boldsymbol{x},t) \to \boldsymbol{E}(t)$$

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If time reversal symmetry:

$\boldsymbol{J}_{free}(\omega)$	$\boldsymbol{P}(\omega)$
diverges as $\omega \to 0$	finite as $\omega \to 0$
"intraband"	"interband"

If not, more complicated...

SciPost Phys. 14, 058 (2023)

Linear response

Beyond the "long wavelength" limit

$$\boldsymbol{E}(\boldsymbol{x}, t)$$

$$\boldsymbol{B}(\boldsymbol{x}, t)$$

Linear response

Beyond the "long wavelength" limit

$$\boldsymbol{E}(\boldsymbol{x},t)$$

$$\boldsymbol{B}(\boldsymbol{x},t)$$

$$\boldsymbol{J}(\boldsymbol{x},\omega) = \int \frac{d\boldsymbol{q}}{(2\pi)^3} \boldsymbol{J}(\boldsymbol{q},\omega) e^{i\boldsymbol{q}\cdot\boldsymbol{x}}$$

Faraday's law: $$\boldsymbol{B}(\boldsymbol{q},\omega) = \frac{c}{\omega}\boldsymbol{q} \times \boldsymbol{E}(\boldsymbol{q},\omega)$$

Linear response

Beyond the "long wavelength" limit

$$\boldsymbol{E}(\boldsymbol{x}, t)$$

$$\boldsymbol{B}(\boldsymbol{x}, t)$$

$$J^i(\boldsymbol{q}, \omega) = \sigma^{ij}(\omega) E^j(\boldsymbol{q}, \omega) + \sigma^{ijk}(\omega) E^j(\boldsymbol{q}, \omega) q^k + \sigma^{ijkl}(\omega) E^j(\boldsymbol{q}, \omega) q^k q^l + ...$$

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magnetoelectric effect,
optical activity,
.....

Linear response

Beyond the "long wavelength" limit

$$\boldsymbol{E}(\boldsymbol{x},t)$$

$$\boldsymbol{B}(\boldsymbol{x},t)$$

$$\mathsf{F}^{ij}(\boldsymbol{x},t)=\frac{1}{2}\left(\frac{\partial E^i(\boldsymbol{x},t)}{\partial x^j}+\frac{\partial E^j(\boldsymbol{x},t)}{\partial x^i}\right)$$

$$J^i(\boldsymbol{q},\omega)=\sigma^{ij}(\omega)E^j(\boldsymbol{q},\omega)+\sigma^{ijk}(\omega)E^j(\boldsymbol{q},\omega)q^k+\sigma^{ijkl}(\omega)E^j(\boldsymbol{q},\omega)q^kq^l+\ldots..$$

initial focus on topologically trivial insulators

Electric dipole moment per unit volume	←	$\boldsymbol{B}$ $\quad$ F
Magnetic dipole moment per unit volume	←	$\boldsymbol{E}$
Electric quadrupole moment per unit volume	←	$\boldsymbol{E}$

Phys. Rev. Research 2, 033126 (2020)
Phys. Rev. Research 2, 043110 (2020)

Linear response

Beyond the "long wavelength" limit

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Phys. Rev. Research 2, 033126 (2020)
Phys. Rev. Research 2, 043110 (2020)

Extension to include electron spin
Phys. Rev. B106, 085413 (2022)

Linear response

Beyond the "long wavelength" limit

$\boldsymbol{E}(\boldsymbol{x},t)$

$\boldsymbol{B}(\boldsymbol{x},t)$

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gauge dependent

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gauge independent

gauge dependent

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Electric dipole moment per unit volume ← $\boldsymbol{B}$ F

Magnetic dipole moment per unit volume ← $\boldsymbol{E}$

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Phys. Rev. Research 2, 033126 (2020)
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many more contributions!

Linear response

Beyond the "long wavelength" limit

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many more contributions!

almost done

gauge independent

Linear response

Beyond the "long wavelength" limit

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many more contributions!

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gauge independent

From $\omega \to 0$ *limit extract the static magnetic susceptibility*

Linear response

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many more contributions!

almost done

gauge independent

From $\omega \to 0$ *limit extract the static magnetic susceptibility*

Still an open issue in the literature, even for insulators in the independent particle approximation

arXiv: 2306.03820

Nonlinear response

...work in progress....

Including quantum optical effects

…work in progress….

Overview
The story for molecules and atoms
Generalizing to condensed matter
Some results
Perspective

Why bother?

$$\rho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{j}(\boldsymbol{x},t) = \frac{\partial \boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla \times \boldsymbol{m}(\boldsymbol{x},t) + \boldsymbol{j}_{free}(\boldsymbol{x},t)$$

Why not just work with microscopic charge and current densities?

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Why not just work with microscopic charge and current densities?

Well, it's just interesting to see if it can be done…

Why bother?

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Why not just work with microscopic charge and current densities?

It leads to describing the interaction of light through

$$\boldsymbol{E}(\boldsymbol{x},t),\ \boldsymbol{B}(\boldsymbol{x},t)$$

rather than

$$\phi(\boldsymbol{x},t),\ \boldsymbol{A}(\boldsymbol{x},t)$$

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Why not just work with microscopic charge and current densities?

It leads to describing the interaction of light through

$$\boldsymbol{E}(\boldsymbol{x},t),\ \boldsymbol{B}(\boldsymbol{x},t)$$

rather than

$$\phi(\boldsymbol{x},t),\ \boldsymbol{A}(\boldsymbol{x},t)$$

can avoid certain technical problems that arise with minimal coupling calculations

Why bother?

$$\rho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{j}(\boldsymbol{x},t) = \frac{\partial\boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{m}(\boldsymbol{x},t) + \boldsymbol{j}_{free}(\boldsymbol{x},t)$$

Why not just work with microscopic charge and current densities?

once macroscopic fields are calculated from spatial averaging, it is easy to extract multipole expansions:

$\boldsymbol{P}(x,t) =$ electric dipole term + electric quadrupole term + ...

$\boldsymbol{M}(x,t) =$ magnetic dipole term + ...

Why bother?

$$\rho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{p}(\boldsymbol{x},t) + \rho_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{j}(\boldsymbol{x},t) = \frac{\partial\boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{m}(\boldsymbol{x},t) + \boldsymbol{j}_{free}(\boldsymbol{x},t)$$

Why not just work with microscopic charge and current densities?

With "lattice site polarizations and magnetizations" introduced, it is possible to take the "molecular crystal" limit of crystals and see how the response is affected by the ability of electrons to "move through" the lattice.

$$\varrho(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{P}(\boldsymbol{x},t) + \varrho_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{J}(\boldsymbol{x},t) = \frac{\partial \boldsymbol{P}(\boldsymbol{x},t)}{\partial t} + c\nabla \times \boldsymbol{M}(\boldsymbol{x},t) + \boldsymbol{J}_{free}(\boldsymbol{x},t)$$

$$\varrho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{P}(\boldsymbol{x},t) + \varrho_{free}(\boldsymbol{x},t)$$
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$$\boldsymbol{P}'(\boldsymbol{x},t) = \boldsymbol{P}(\boldsymbol{x},t) + \nabla\times\boldsymbol{a}(\boldsymbol{x},t) + \boldsymbol{C}(\boldsymbol{x},t)$$
$$\boldsymbol{M}'(\boldsymbol{x},t) = \boldsymbol{M}(\boldsymbol{x},t) - \frac{1}{c}\frac{\partial\boldsymbol{a}(\boldsymbol{x},t)}{\partial t} + \nabla b(\boldsymbol{x},t)$$
$$\varrho'_{free}(\boldsymbol{x},t) = \varrho_{free}(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{C}(\boldsymbol{x},t)$$
$$\boldsymbol{J}'_{free}(\boldsymbol{x},t) = \boldsymbol{J}_{free}(\boldsymbol{x},t) - \frac{\partial\boldsymbol{C}(\boldsymbol{x},t)}{\partial t}$$

$$\varrho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{P}(\boldsymbol{x},t) + \varrho_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{J}(\boldsymbol{x},t) = \frac{\partial\boldsymbol{P}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{M}(\boldsymbol{x},t) + \boldsymbol{J}_{free}(\boldsymbol{x},t)$$

$$\boldsymbol{P}'(\boldsymbol{x},t) = \boldsymbol{P}(\boldsymbol{x},t) + \nabla\times\boldsymbol{a}(\boldsymbol{x},t) + \boldsymbol{C}(\boldsymbol{x},t)$$

$$\boldsymbol{M}'(\boldsymbol{x},t) = \boldsymbol{M}(\boldsymbol{x},t) - \frac{1}{c}\frac{\partial\boldsymbol{a}(\boldsymbol{x},t)}{\partial t} + \nabla b(\boldsymbol{x},t)$$

$$\varrho'_{free}(\boldsymbol{x},t) = \varrho_{free}(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{C}(\boldsymbol{x},t)$$

$$\boldsymbol{J}'_{free}(\boldsymbol{x},t) = \boldsymbol{J}_{free}(\boldsymbol{x},t) - \frac{\partial\boldsymbol{C}(\boldsymbol{x},t)}{\partial t}$$

$$\varrho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{P}'(\boldsymbol{x},t) + \varrho'_{free}(\boldsymbol{x},t)$$

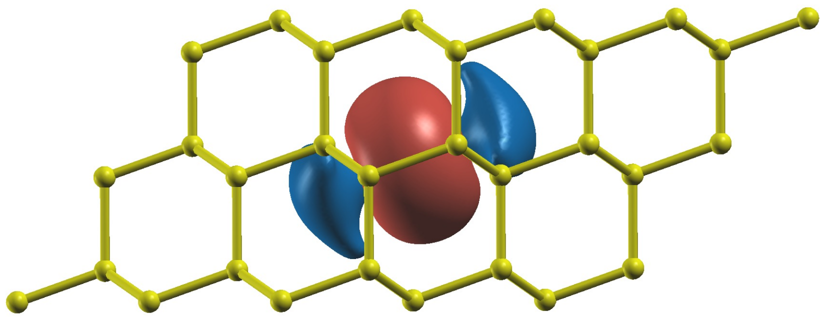
$$\boldsymbol{J}(\boldsymbol{x},t) = \frac{\partial\boldsymbol{P}'(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{M}'(\boldsymbol{x},t) + \boldsymbol{J}'_{free}(\boldsymbol{x},t)$$

$$\varrho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{P}(\boldsymbol{x},t) + \varrho_{free}(\boldsymbol{x},t)$$
$$\boldsymbol{J}(\boldsymbol{x},t) = \frac{\partial\boldsymbol{P}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{M}(\boldsymbol{x},t) + \boldsymbol{J}_{free}(\boldsymbol{x},t)$$

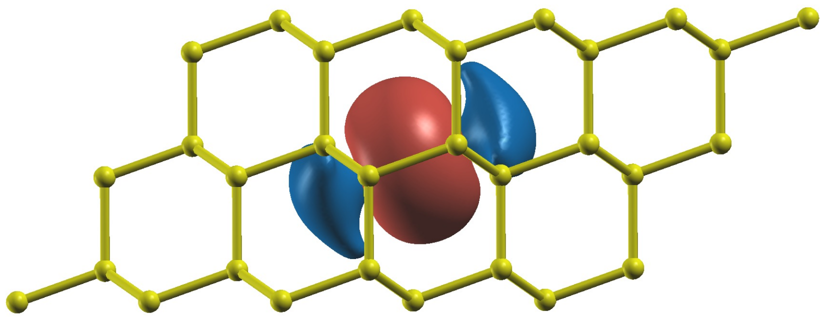
$$\boldsymbol{P}'(\boldsymbol{x},t) = \boldsymbol{P}(\boldsymbol{x},t) + \nabla\times\boldsymbol{a}(\boldsymbol{x},t) + \boldsymbol{C}(\boldsymbol{x},t)$$
$$\boldsymbol{M}'(\boldsymbol{x},t) = \boldsymbol{M}(\boldsymbol{x},t) - \frac{1}{c}\frac{\partial\boldsymbol{a}(\boldsymbol{x},t)}{\partial t} + \nabla b(\boldsymbol{x},t)$$
$$\varrho'_{free}(\boldsymbol{x},t) = \varrho_{free}(\boldsymbol{x},t) + \nabla\cdot\boldsymbol{C}(\boldsymbol{x},t)$$
$$\boldsymbol{J}'_{free}(\boldsymbol{x},t) = \boldsymbol{J}_{free}(\boldsymbol{x},t) - \frac{\partial\boldsymbol{C}(\boldsymbol{x},t)}{\partial t}$$

$$\varrho(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{P}'(\boldsymbol{x},t) + \varrho'_{free}(\boldsymbol{x},t)$$
$$\boldsymbol{J}(\boldsymbol{x},t) = \frac{\partial\boldsymbol{P}'(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{M}'(\boldsymbol{x},t) + \boldsymbol{J}'_{free}(\boldsymbol{x},t)$$

Use this opportunity!

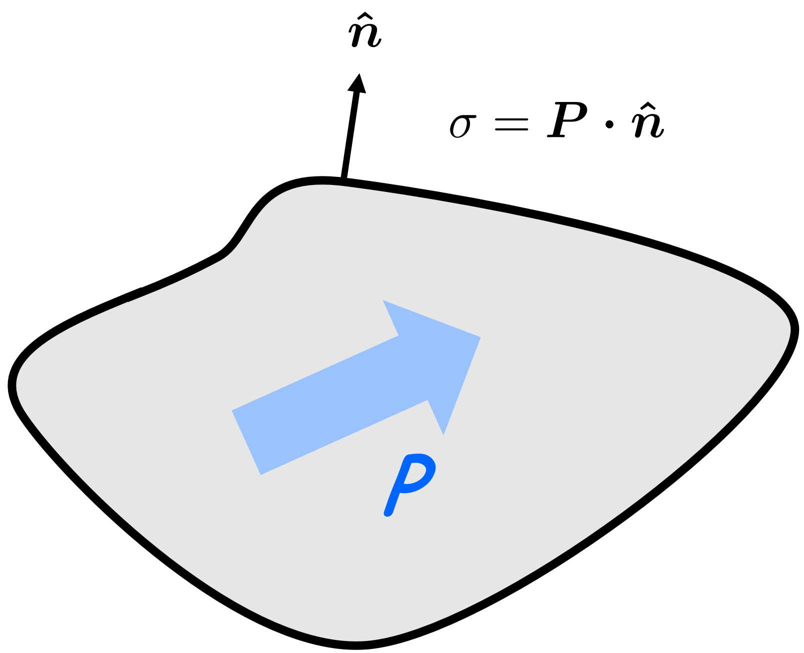


Choose the Wannier functions to help in approximate descriptions of the electron-electron interaction



Choose the Wannier functions to help in approximate descriptions of the electron-electron interaction

Use relations between bulk and surface quantities to explore best definitions for bulk quantities and surface quantities



"Optical alchemy"