On the Verdet constant and Faraday rotation for graphene-like materials, QGRAPH Network meeting.

Mikkel H. Brynildsen

Department of Mathematics, Aalborg University.
Joint work with Horia Cornean, Aalborg University.

December 9, 2012

Aalborg University

- M. H. Brynildsen[1] and H. D. Cornean[1]: "On the Verdet constant and Faraday rotation for graphene-like materials." arxiv.org/abs/1112.2613
- Jesper Goor Pedersen[2], Mikkel H. Brynildsen[1], Horia D. Cornean[1], and Thomas Garm Pedersen[2,3]: "Optical Hall conductivity in bulk and nanostructured graphene beyond the Dirac approximation" (submitted)

1: Department of Mathematical Sciences, Aalborg University,
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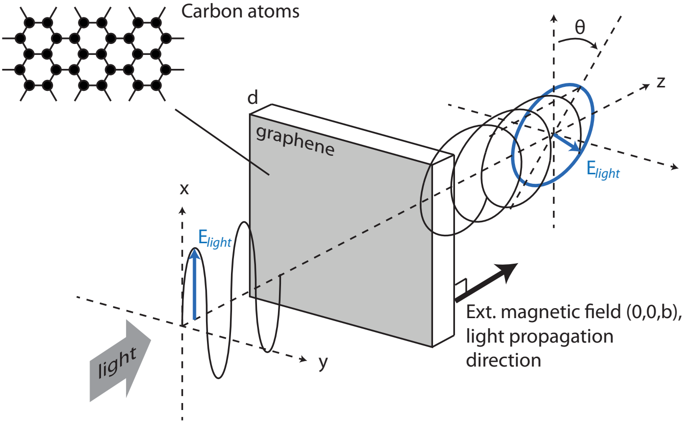
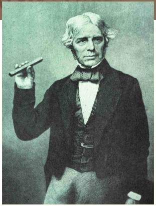
3: Center for Nanostructured Graphene (CNG), Aalborg University

Faraday effect



- Polarization changes as the light travels through the layer. ($\theta = 0,1$ radian rotation reported in single layer graphene).
- Linear regime: $\theta_{linear} = Vbd$, V is the Verdet constant.



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- →1866: Marcel Emile Verdet, dependence of the Faraday effect on magnetic field-strength.
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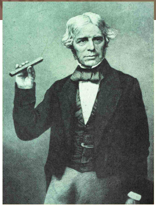

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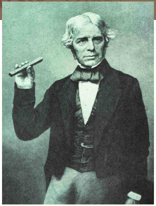

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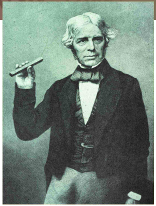

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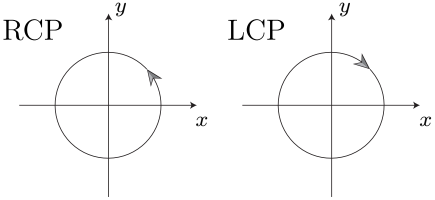
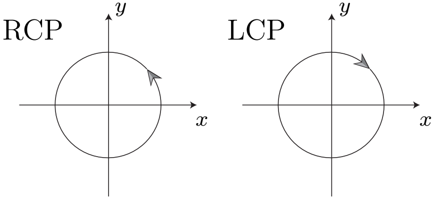
Optical activity: phase differences for circular polarized waves of opposite handedness in transmitted light-wave.



For a specific point in space, write
$\boldsymbol{E}(t) = \hat{x}E\cos(\omega t - \psi_x) + \hat{y}E\cos(\omega t - \psi_y)$.

Optical activity: phase differences for circular polarized waves of opposite handedness in transmitted light-wave.



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$\boldsymbol{E}(t) = \hat{x}E\cos(\omega t - \psi_x) + \hat{y}E\cos(\omega t - \psi_y)$.
If $\psi_y - \psi_x = \pi/2$ then $\boldsymbol{E}(t) = \hat{x}E\cos(\omega t - \psi_x) + \hat{y}E\sin(\omega t - \psi_x)$ right-hand circularly polarized (RCP).

$$\boldsymbol{E}_{RCP}(t) = \mathrm{Re}\left\{\mathrm{E}(\hat{\mathrm{x}} - \mathrm{i}\hat{\mathrm{y}})\mathrm{e}^{\mathrm{i}(\omega \mathrm{t} - \psi_{\mathrm{x}})}\right\}$$

Faraday effect

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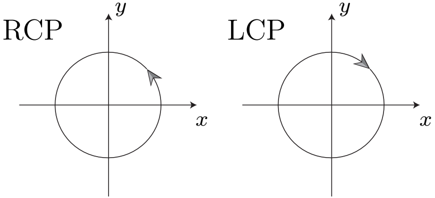

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If $\psi_y - \psi_x = -\pi/2:$ left-hand circularly polarized (LCP).

$$\boldsymbol{E}_{LCP}(t) = \mathrm{Re}\left\{\mathrm{E}(\hat{\mathrm{x}} + \mathrm{i}\hat{\mathrm{y}})\mathrm{e}^{\mathrm{i}(\omega \mathrm{t} - \psi_{\mathrm{x}})}\right\}$$

Maxwell's equations in linear media, Ohm's law, zero charge density.

$$\nabla \times \boldsymbol{B} = \mu\overline{\overline{\sigma}}\boldsymbol{E} + \mu\epsilon\frac{\partial \boldsymbol{E}}{\partial t}, \qquad \nabla \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t},$$
$$\nabla \cdot \boldsymbol{E} = 0, \qquad \nabla \cdot \boldsymbol{B} = 0.$$

apply curl and suppose $\boldsymbol{E}(x,y,z,t) = \mathrm{Re}\ \{\boldsymbol{E}_0 \exp \mathrm{i}(\omega \mathrm{t} - \mathrm{kz})\}$

$$\nabla(\nabla \cdot \boldsymbol{E}) - \nabla^2 \boldsymbol{E} = -\frac{\partial}{\partial t}(\mu\overline{\overline{\sigma}}\boldsymbol{E} + \mu\epsilon\frac{\partial \boldsymbol{E}}{\partial t})$$
$$k^2\boldsymbol{E} = -i\omega\mu\overline{\overline{\sigma}}\boldsymbol{E} + \omega^2\mu\epsilon\boldsymbol{E}.$$

RCP $\boldsymbol{E}_0 = E(\hat{x} - i\hat{y})$, LCP $\boldsymbol{E}_0 = E(\hat{x} + i\hat{y})$ disregard z-components
$\sigma_{ij}^{2D} = d\sigma_{ij}^{3D}$

$$k_{\pm}^2 E \begin{bmatrix} 1 \\ \pm i \end{bmatrix} = \omega^2\mu\epsilon(1 - \frac{i}{\epsilon\omega}) \begin{bmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{bmatrix} E \begin{bmatrix} 1 \\ \pm i \end{bmatrix}$$

Faraday effect

$$k_\pm^2 = (\eta_\pm + i\kappa_\pm)^2 = \omega^2 \mu\epsilon (1 \pm \frac{\sigma_{xy}}{\epsilon\omega} - \frac{i\sigma_{xx}}{\epsilon\omega})$$

Suppose expansions of σ_{ij} (we show this for $\sigma_{xy}(b)$ in our model):

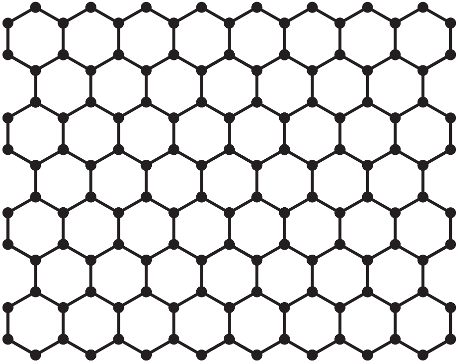
$$\sigma_{xx}(b) = \sigma_{xx}^{[0]} + \sigma_{xx}^{[1]} b + \sigma_{xx}^{[2]} b^2 + \ldots$$
$$\sigma_{xy}(b) = \sigma_{xy}^{[1]} b + \sigma_{xy}^{[2]} b^2 + \ldots$$

Transmitted beam: phase difference between RCP and LCP is determined by difference in the real part of the wave vectors for circular polarized waves of opposite handedness

$$\eta_\pm(b) = f(\sigma_{xx}(b), \pm\sigma_{xy}(b))$$
$$(\eta_+(b) - \eta_-(b)) \approx \partial_2 f(\sigma_{xx}^{[0]}, 0)\ \sigma_{xy}^{[1]}\ b$$
$$\sigma_{xy}^{[1]} \leftrightarrow \text{Verdet constant}$$

Detail: $\sigma_{xy} = -\sigma_{yx}$ from symmetries. We will actually find σ_{yx}.

Honeycomb lattice



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Graphene

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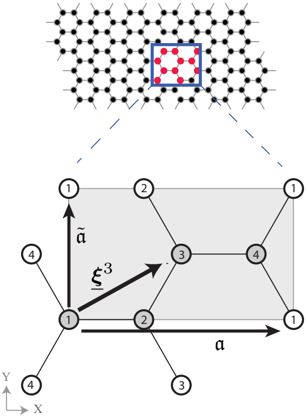
- I. Crassee and J. Levallois, A. L. Walter, M. Ostler A. Bostwick, E. Rotenberg, T. Seyller, D. van der Marel, A. B. Kuzmenko: "Giant Faraday rotation in single- and multilayer graphene", 2010.

A rectangular lattice crystal structure of graphene

$$\begin{aligned}\Gamma &= \text{Bravais lattice} \\ &= \{m\mathfrak{a} + n\tilde{\mathfrak{a}}\}_{m,n\in\mathbb{Z}} \subset \text{xy-plane} \;. \\ \mathfrak{B} &= \text{basis} \;\; = \{\underline{\boldsymbol{\xi}}^1, \ldots, \underline{\boldsymbol{\xi}}^4\} \subset \text{xy-plane} \;, \\ \Omega &= \text{unit cell} \;\; = \{\theta_1\mathfrak{a} + \theta_2\tilde{\mathfrak{a}}\}_{\theta_i\in[0,1]}\end{aligned}$$



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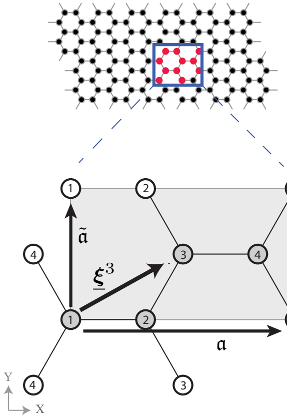
\Gamma^* &= \text{dual lattice} = \{m\mathfrak{a}^* + n\tilde{\mathfrak{a}}^*\}_{m,n\in\mathbb{Z}} \\
&\qquad \mathfrak{a}\cdot\mathfrak{a}^* = \tilde{\mathfrak{a}}\cdot\tilde{\mathfrak{a}}^* = 2\pi, \\
&\qquad \tilde{\mathfrak{a}}\cdot\mathfrak{a}^* = \tilde{\mathfrak{a}}\cdot\mathfrak{a}^* = 0. \\
\Omega^* &= \text{First Brilluoin zone} \\
&= \{\boldsymbol{k}\in\mathbb{R}^2 \ : \ \|\boldsymbol{k}\| \le \|\boldsymbol{k} - \gamma*\|, \ \gamma^*\in\Gamma^*\}
\end{aligned}$$



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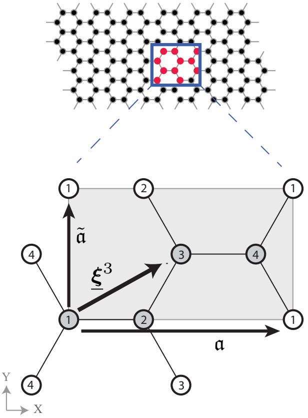
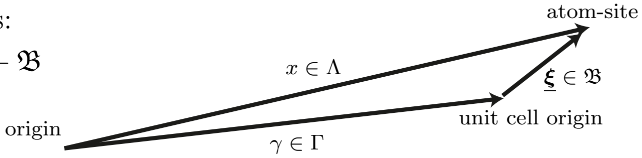


Family of atom-sites:

$$\Lambda = \Gamma + \mathfrak{B}$$



QM model, Peierls substitution

Configuration space is $\ell^2(\Lambda)$ (Hilbert space of one-electron states).
Zero-field Hamiltonian H_0 with int. kernel t:

$$t(x,x') = \langle \delta_x, H_0\, \delta_{x'} \rangle\,, \quad (x,x' \in \Lambda)$$

(t is the "Hopping matrix") Nearest-neighbour interactions only:

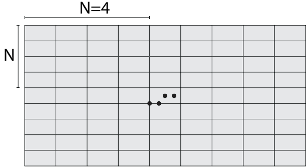
$$t(x,x') = 1, \quad \text{if } x \text{ and } x' \text{ are neighbours, zero otherwise.}$$

Peierls substitution; magnetic Hamiltonian H_b:

$$H_b(x,x') := e^{ib\varphi(x,x')}\, t(x,x'), \quad b\,\varphi(x,x') = \int_x^{x'} a(s)ds = b\,\frac{x'_x x_y - x'_y x_x}{2}.$$

Symmetric gauge:

$$a(r_1,r_2) := \frac{b}{2}\,(-x_2,x_1)\,, \quad Curl\ a(x) = B(x) = b.$$



Central region Λ_N of $(2N+1)^2$ unit cells

$$\Lambda_N := \{(\gamma + \underline{x}) \in \Lambda \ : \ \gamma = m\mathfrak{a} + n\tilde{\mathfrak{a}},\ |m| \leq N, |n| \leq N, \underline{x} \in \mathfrak{B}\}. \tag{1}$$

Dirichlet Boundary Conditions:

$$\chi_N = \text{the characteristic function of } \Lambda_N \tag{2}$$

$$H_{b,N} = \chi_N H_b \chi_N \tag{3}$$

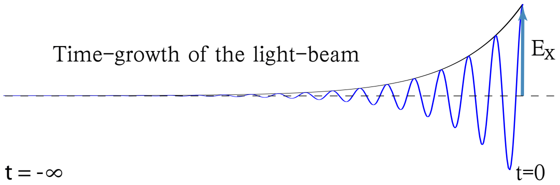
(notation: subscript N)

Light beam, time-dependent perturbation

$\hat{X}$ and $\hat{Y}$ are the position operators; $\hat{X}\delta_x = x_1\delta_x$, $\hat{Y}\delta_x = x_2\delta_x$.
Perturbation by incident polarized light-beam with complex frequency $\omega = \omega_0 - \eta i$, $\mathrm{Re}\,(\omega) = \omega_0 > 0$, $\mathrm{Im}\,(\omega) = -\eta < 0$:

$$\begin{aligned} H_{E,b}(t) &= H_b + V_E(t), \\ V_E(t) &= Ee^{\eta t}\cos(\omega_0 t)\hat{X} \end{aligned} \tag{4}$$

$$\boldsymbol{E}(t) = \left(\mathrm{Re}\,\left[\mathrm{Ee}^{\mathrm{i}\omega\mathrm{t}}\right], 0\right) = (Ee^{\eta t}\cos(\omega_0 t), 0) \tag{5}$$



Small η equals slow "turning on". $\eta \to 0$ is the adiabatic limit.

Kubo formalism, y-current density, $\sigma_{yx,N}$

Fermi-Dirac distribution

$$f(z) = \frac{1}{e^{\beta(z-\mu)} + 1} \tag{6}$$

Density operator satisfies:

$$i\frac{\mathrm{d}\varrho_N}{\mathrm{d}t}(t) = [H_{E,b,N}, \varrho_N(t)], \quad \lim_{t\to-\infty} \varrho_N(t) = f(H_{b,N}). \tag{7}$$

Current operator in the y-direction:

$$j_{y,N} = i[H_{E,b,N}, \hat{Y}_N] = i[H_{b,N}, \hat{Y}_N] \tag{8}$$

Current density in the y-direction at time $t = 0$:

$$J_{y,N}(E) = \frac{1}{|\Lambda_N|} \operatorname{Tr}\{\varrho_N(t=0)\, j_{y,N}\} = E\, \sigma_{yx,N}(b) + \mathcal{O}(E^2). \tag{9}$$

(Defines $\sigma_{yx,N}(b)$).

Kubo formalism, y-current density, $\sigma_{yx,N}$

Schrödinger picture:

$$H_N = H_{b,N} + V_{E,N}(t), \qquad \text{operator } A_N \text{ (time-independent)}$$

Interaction picture:

$$\tilde{A}_N = e^{itH_{b,N}} A_N e^{-itH_{b,N}} \qquad i\frac{\mathrm{d}}{\mathrm{d}t}\tilde{A}_N = [\tilde{A}_N, H_{b,N}].$$

(at $t = 0$, $\tilde{A}_N = A_N$.) Density operator in interaction picture:

$$i\frac{\mathrm{d}}{\mathrm{d}t}\tilde{\varrho}_N(t) = [\tilde{V}_{E,N}(t), \tilde{\varrho}(t)]$$

($\tilde{\varrho}_{E,N}(t_0) = \exp(it_0 H_{b,N})\varrho_{E,N}(t_0)\exp(-it_0 H_{b,N}) \to f(H_{b,N})$ as $t_0 \to -\infty$.)

Kubo formalism, y-current density, $\sigma_{yx,N}$

$$< J_{2,b,N} >(t=0) = \mathrm{Tr}\left\{ \varrho_{N,E}(0) \frac{1}{|\Lambda_N|} j_{2,N,b} \right\} = \mathrm{Tr}\left\{ \tilde{\varrho}_{N,E}(0) \frac{1}{|\Lambda_N|} j_{2,N,b} \right\}$$

$$\begin{aligned}
\tilde{\varrho}_{N,E}(0) &= \lim_{t_0 \to -\infty} \left\{ \tilde{\varrho}_{N.E}(t_0) + \int_{t_0}^{0} \left(\frac{\mathrm{d}}{\mathrm{d}s} \tilde{\varrho}_{N.E}(s) \right) \, ds \right\} \\
&= f(H_{b,N}) + \int_{-\infty}^{0} \left(-i[\tilde{V}_{E,N}(s), \tilde{\varrho}_{E,N}(s)] \right) \, ds \\
&= f(H_{b,N}) - iE \int_{-\infty}^{0} e^{i\omega s} \left([\tilde{\hat{X}}_{1,E,N}(s), f(H_{b,N})] \right) \, ds + \mathcal{O}(E^2)
\end{aligned}$$

main steps..

$$\frac{\mathrm{d}}{\mathrm{d}t} \left(e^{itH_{N,b}} \hat{X}_{1,N} e^{-itH_{N,b}} \right) = e^{itH_{N,b}} j_{1,b,N} e^{-itH_{N,b}}$$

$$e^{-is(H_{N,b}-\omega)} f(H_{b,N}) = \frac{1}{2\pi} \int_{\mathscr{C}} e^{-is(z-\omega)} f(z) (H_{b,N} - z)^{-1} \, dz$$

Main result 1

$$\tilde{T}_N(z) := (H_{b,N} - z + \omega)^{-1} j_{x,N} (H_{b,N} - z)^{-1} j_{y,N},$$

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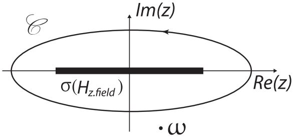
Main result 1: Thermodynamic, adiabatic limit:

$$\sigma_{yx}(b) := \lim_{\eta \to 0} \lim_{N \to \infty} \left(\frac{\eta}{(\eta^2 + \omega_0^2)|\Lambda_N|} \mathrm{Tr}\left\{ i[j_{y,N}, \hat{X}_N] f(H_{b,N}) \right\} + \mathrm{Re} \frac{1}{2\pi\omega|\Lambda_N|} \int_{\mathscr{C}} \mathrm{dz}\, \mathrm{f(z)}\, \mathrm{Tr}\left\{ \tilde{\mathrm{T}}_{\mathrm{N}}(\mathrm{z}) + \tilde{\mathrm{T}}_{\mathrm{N}}(\mathrm{z} + \omega) \right\} \right)$$



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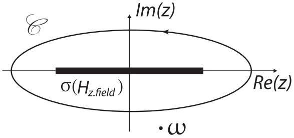
$$+ \mathrm{Re}\, \frac{1}{2\pi\omega|\Lambda_N|} \int_{\mathscr{C}} \mathrm{dz}\, \mathrm{f(z)}\, \mathrm{Tr}\left\{ \tilde{\mathrm{T}}_\mathrm{N}(\mathrm{z}) + \tilde{\mathrm{T}}_\mathrm{N}(\mathrm{z} + \omega) \right\} \Bigg)$$

$\downarrow$

Geometric perturbation,

$\downarrow$.



$$= \mathrm{Re}\, \frac{1}{2|\Omega|\pi\omega_0} \int_{\mathscr{C}} \mathrm{dz}\, \mathrm{f(z)} \sum_{\underline{\mathrm{x}} \in \Omega} \{ [(\mathrm{H_b} - \mathrm{z} + \omega_0)^{-1} \mathrm{j_x} (\mathrm{H_b} - \mathrm{z})^{-1} \mathrm{j_y}](\underline{\mathrm{x}}, \underline{\mathrm{x}})$$
$$+ (H_b - z)^{-1} j_x (H_b - z - \omega_0)^{-1} j_y](\underline{x}, \underline{x}) \}$$

Main result 1

Problem: magnetic resolvents...Naive perturbation will not work:

$$H_b - z = \left[I + (H_b - H_0)(H_0 - z)^{-1}\right](H_0 - z) \tag{10}$$

(10) invertible for $z \in \rho(H_0)$, weak magnetic field, if

$$\|H_b - H_0\| \to 0, \quad b \to 0.$$

Naive pertubation theory in the linear regime:

$$[H_b - H_0](x, x') = (e^{ib\varphi(x,x')} - 1)t(x, x') \approx ib\varphi(x, x')t(x, x')$$

does not work; no matter how close to zero b is!, $\varphi(x, x')$ is unbounded. workaround, why not contruct $S(b), K(b)$ operators s.t.

$$(H_b - z)S(b) = I + K(b), \qquad \|K(b)\| < 1 \text{ when } 0 \leq b < b_K \text{ ?} \tag{11}$$

Magnetic perturbation

$$S_b(x,x') := e^{ib\varphi(x,x')}(H_0 - z)^{-1}(x,x')$$

$$K_b(x,x') := e^{ib\varphi(x,x')} \sum_{x''\in\Lambda} \left(e^{ib[FL]} - 1\right) H_0(x,x'')S_0(x'',x')$$

$$[FL] = \varphi(x,x'') + \varphi(x'',x') + \varphi(x',x)$$

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$$K_b(x,x') := e^{ib\varphi(x,x')} \sum_{x''\in\Lambda} \left(e^{ib[FL]} - 1\right) H_0(x,x'')S_0(x'',x')$$

$$[FL] = \varphi(x,x'') + \varphi(x'',x') + \varphi(x',x)$$

$$(H_b - z)S_b = I + K_b, \qquad \|K_b\| < 1 \text{ when } 0 \le b < b_K \tag{12}$$

$$(H_b - z)^{-1} = S_b(z) - S_b(z)K_b(z) + (H_b - z)^{-1}K_b(z)^2 \tag{13}$$

$$\sup_{z\in\mathscr{C}}\{\|K_b(z)\|\} \le b\; const. \tag{14}$$

$$\sup_{z\in\mathscr{C}}|[(H_b - z)^{-1}K_b(z)^2](x,x')| \le b^2\; const. \tag{15}$$

Magnetic perturbation

$$S_b(x,x') := e^{ib\varphi(x,x')}(H_0 - z)^{-1}(x,x')$$

$$K_b(x,x') := e^{ib\varphi(x,x')} \sum_{x''\in\Lambda} \left(e^{ib[FL]} - 1\right) H_0(x,x'')S_0(x'',x')$$

$$[FL] = \varphi(x,x'') + \varphi(x'',x') + \varphi(x',x)$$

$$(H_b - z)S_b = I + K_b, \qquad \|K_b\| < 1 \text{ when } 0 \le b < b_K \tag{12}$$

$$(H_b - z)^{-1} = S_b(z) - S_b(z)K_b(z) + (H_b - z)^{-1}K_b(z)^2 \tag{13}$$

$$\sup_{z\in\mathscr{C}}\{\|K_b(z)\|\} \le b\; const. \tag{14}$$

$$\sup_{z\in\mathscr{C}}|[(H_b - z)^{-1}K_b(z)^2](x,x')| \le b^2\; const. \tag{15}$$

- H_0, $(H_0 - z)^{-1}$ periodic in Γ
- Bloch-Floquet decomposition, k-space representation.
- b-dependence only in flux terms.

Bloch-Floquet decomposition

$\ell^2(\Lambda)$ unitarily equivalent with $L^2(\Omega^*) \otimes \mathbb{C}^4$, the zero field Hamiltonian (hopping matrix) is a fibered operator

$$U : \ell^2(\Lambda) \to \mathscr{H}_F, \quad (U\psi)(k, \underline{x}) := \frac{1}{\sqrt{|\Omega^*|}} \sum_{\gamma \in \Gamma} e^{-ik(\underline{x}+\gamma)} \psi(\underline{x} + \gamma) \tag{16}$$

$$U H_0 U^{-1} = \int_{\Omega^*}^{\oplus} dk \; H(k), \tag{17}$$

Bloch-Floquet decomposition formulae:

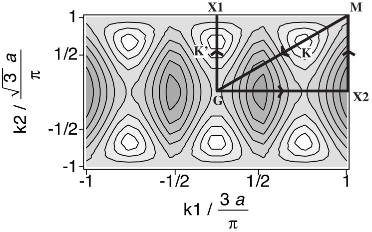
$$\begin{aligned} H(\underline{x}, \underline{x}'; k) &= \frac{1}{|\Omega^*|} \sum_{\gamma \in \Gamma} e^{-ik\cdot(\underline{x}+\gamma-\underline{x}')} t(\underline{x} + \gamma, \underline{x}'). \\ t(x, x') &= \frac{1}{|\Omega^*|} \int_{\Omega^*} dk \; e^{ik\cdot(x-x')} H(\underline{x}, \underline{x}'; k), \\ x = \underline{x} + \gamma, &\qquad x' = \underline{x}' + \gamma'. \end{aligned} \tag{18}$$

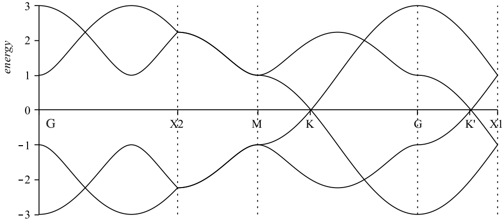
$H(k)$ are 4×4 matrices. (Generally $\#\mathfrak{B} \times \#\mathfrak{B}$ matrices).

$H(k_x, k_y) =$ zero field, nearest-neighbour Hamiltonian fiber, for $k = (k_x, k_y) \in \Omega^*$:

$$\begin{bmatrix} 0 & e^{ik_x a} & 0 & 2e^{-\frac{ik_x a}{2}} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) \\ e^{-ik_x a} & 0 & 2e^{\frac{ik_x a}{2}} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) & 0 \\ 0 & 2e^{-\frac{ik_x a}{2}} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) & 0 & e^{ik_x a} \\ 2e^{\frac{ik_x a}{2}} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) & 0 & e^{-ik_x a} & 0 \end{bmatrix}.$$

$a =$ nearest neighbour atom-atom distance.


X1
M
K'
K
G
X2
1
1/2
-1/2
-1
-1
-1/2
1/2
1
k2 / $\frac{\sqrt{3}\,a}{\pi}$
k1 / $\frac{3\,a}{\pi}$


energy
3
2
1
0
−1
−2
−3
G
X2
M
K
G
K'
X1

Short notation on this slide:

$$G(k_x,k_y,z) = (H(k_x,k_y)-z)^{-1}, \qquad \tilde{G} = (H-z+\omega_0)^{-1}$$

$$H_x(k_x,k_y) = \frac{\partial H}{\partial k_x}(k_x,k_y), \quad H_y(k_x,k_y) = \frac{\partial H}{\partial k_y}(k_x,k_y), \text{ etc.}$$

$$H_{ij}(k_x,k_y) = \frac{\partial^2 H}{\partial k_i \partial k_j}(k_x,k_y), \quad i,j \in \{x,y\}$$

Put

$$\begin{aligned} T(z) = \Big\{ & \tilde{G}_x H_x G_y - \tilde{G}_y H_x G_x \\ & + \tilde{G}[H_x G(H_x G_y - H_y G_x) + (H_y \tilde{G}_x - H_y \tilde{G}_x) H_x G] \Big\} H_y \\ & + \ \Big\{ (\tilde{G}_x H_{xy} - \tilde{G}_y H_{xx}) G + \tilde{G}(H_{xx} G_y - H_{yx} G_x) \Big\} H_y, \end{aligned}$$

then (second and last main result)

$$\left.\frac{\mathrm{d}\sigma_{yx}(\omega_0)}{\mathrm{d}b}\right|_{b=0} = \mathrm{Re}\ \frac{1}{16\pi^3\omega_0} \int_{\Omega^*} \mathrm{dk} \int_{\mathscr{C}} \mathrm{dz\ i\ f(z)\ Tr}_\Omega \left\{ \mathrm{T(z)} + \mathrm{T(z+\omega_0)} \right\}$$

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