Gauge-Invariant Cumulants and Second Harmonic Generation

Berkeley Quantum Materials Group Meeting

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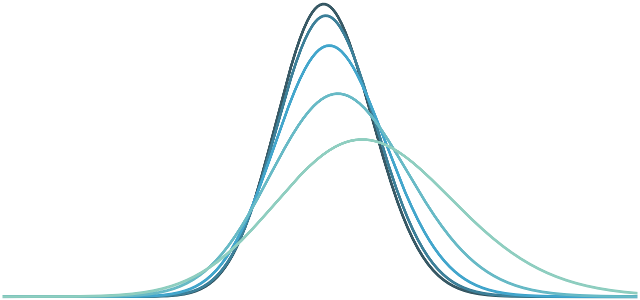
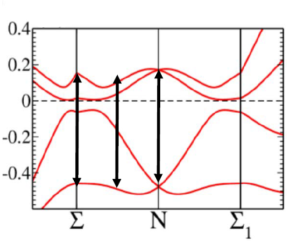


Daniel Eric Parker

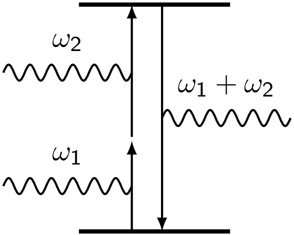
UC Berkeley

Optical Response in Metals

Bands



Diagrams



Formulas

$$
\begin{aligned}
&\mathrm{Re}\,\sigma^{abc}_{\mathrm{SHG}}(-2\omega;\omega,\omega) \\
&= \frac{i\pi e^3}{2\hbar^2\omega^2}\sum_{m,n,p}\int\frac{d^d k}{(2\pi)^d}\Bigg[\\
&\quad \left(v^a_{mn} w^{bc}_{nm} + 2v^a_{mn}\frac{v^b_{np}v^c_{pm} + v^c_{np}v^b_{pm}}{\omega_{mp}+\omega_{np}} \right) f_{mn}\delta_{2\omega-\omega_{nm}} \\
&\quad + \left(w^{ab}_{mn} v^c_{nm} + w^{ac}_{mn} v^b_{nm} \right) f_{mn}\delta(\omega-\omega_{nm}) \\
&\quad + v^a_{mn}\frac{v^b_{np}v^c_{pm} + v^c_{np}v^b_{pm}}{\omega_{pm}+\omega_{pn}}\left(f_{mp}\delta(\omega-\omega_{pm}) - f_{np}\delta(\omega-\omega_{np}) \right)\Bigg]
\end{aligned}
$$

Can we understand optical responses more easily in real space?

Polarization Distribution

- Polarization in a solid:

$$\widehat{P} = e\widehat{X} + q\widehat{X}_{\text{nuc}}; \quad \widehat{X} = \sum_{i \in \text{el}} \hat{x}_i$$

- Assume nuclei are fixed, so $q\widehat{X}_{\text{nuc}} = 0$.
- Much better to work with the **polarization distribution**:

$$p(X) = \left\langle \Psi \middle| \delta(X - \widehat{X}) \middle| \Psi \right\rangle.$$

- This is the probability that the center of charge is exactly at the position X.

W. Kohn. PRL. 1964; I. Souza, T. Wilkens, R. M. Martin. PRB. 2000.

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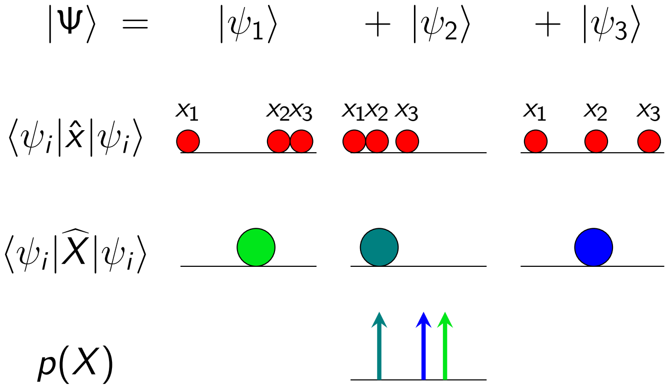

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Gauge-Invariant Cumulants

Calculating the full distribution $p(X)$ is hard.
Instead, we calculate the **gauge-invariant cumulants**:

$$C_1 = \langle X \rangle$$

$$C_2 = \langle X^2 \rangle - \langle X \rangle^2$$

$$C_3 = \langle X^3 \rangle - 3 \langle X^2 \rangle \langle X \rangle + 2 \langle X \rangle^3$$

Gauge-Invariant Cumulants

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$$C_1 = \langle X \rangle \equiv \frac{L}{2\pi} \int dk \, \mathrm{Tr}\left[c_1\right]$$

$$C_2 = \langle X^2 \rangle - \langle X \rangle^2 \equiv \frac{L}{2\pi} \int dk \, \mathrm{Tr}\left[c_2 - c_1^2\right]$$

$$C_3 = \langle X^3 \rangle - 3 \langle X^2 \rangle \langle X \rangle + 2 \langle X \rangle^3 \equiv \frac{L}{2\pi} \int dk \, \mathrm{Tr}\left[c_3 - 3 c_2 c_1 + 2 c_1^3\right]$$

where the c_i's are written in terms of single-particle wavefunctions $|u_{kn}\rangle$ as

$$c_1 \equiv i \langle u_{kn} | \partial_k u_{kn} \rangle \,, \quad c_2 \equiv i^2 \langle u_{kn} | \partial_k^2 u_{kn} \rangle \,, \quad c_3 \equiv i^3 \langle u_{kn} | \partial_k^3 u_{kn} \rangle \,.$$

These are invariant under gauge transformations $|u_{kn}\rangle \to |u_{kn}\rangle \, e^{i\theta_n(k)}$.

I. Souza, T. Wilkens, R. M. Martin. PRB. 2000.

C_1

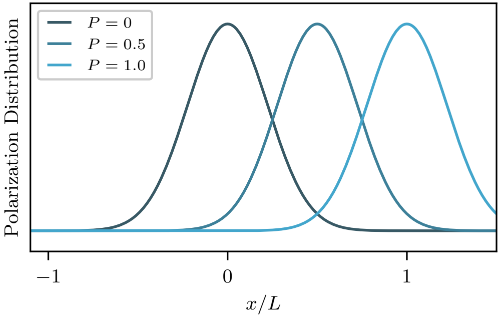
- C_1 is the *mean* of the polarization distribution

$$C_1 = \frac{L}{2\pi} \sum_{n \in \text{occ}} \int dk \; i \langle u_{kn} | \partial_k u_{kn} \rangle \, .$$

- *Berry phase* $A_{kn} = i \langle u_{kn} | \partial_k u_{kn} \rangle$!
- Berry phase theory of polarization:

$$\langle \widehat{P} \rangle = \frac{e}{L} C_1 = \frac{e}{2\pi} \sum_{n \in \text{occ}} \int dk \; A_{kn}$$

- Connection between *geometry* and *optical response*.

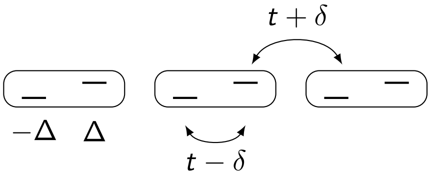


J. Zak. *PRL*. (1989); R.D. King-Smith, D. Vanderbilt. *PRB*. (1993);
R. Resta. *Rev. Mod. Phys.* (1994); R. Resta, D. Vanderbilt. *Phys. of Ferroelectrics*. (2007).

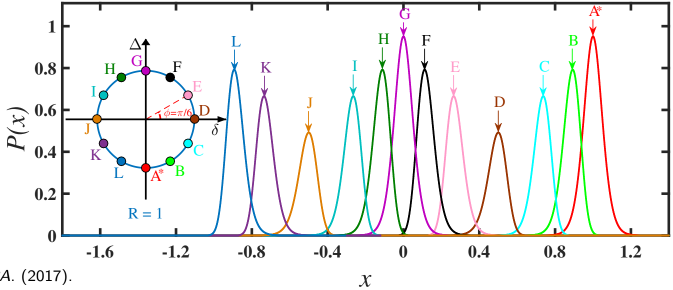
Example: Rice-Mele Model

Rice-Mele is a simple model for ferroelectrics.

$$H_{\text{RM}} = \sum_n (-1)^n \Delta c_n^\dagger c_n + \left(\frac{t}{2} + (-1)^n \frac{\delta}{2}\right) c_n^\dagger c_{n+1} + \text{h.c.}$$



Metallic at $\Delta = \delta = 0$. Circling this point in parameter space pumps charge.



M. Yahyavi, B. Hetényi. *PRA*. (2017).

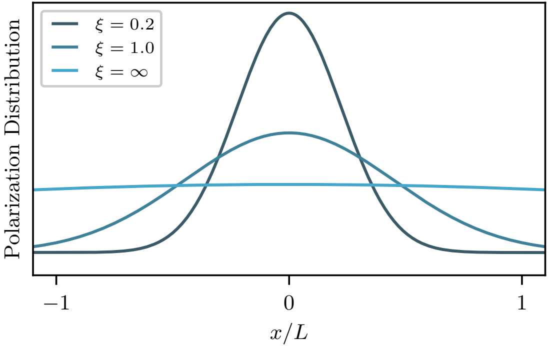
C_2

- C_2 is the *variance*.
- Connected to quantum metric.
- Connected to electron localization

$$\xi = \sqrt{C_2/L}$$

- C_2 diverges for metals where electrons are "free"
- Delocalized electrons are more conductive. Sum rule:

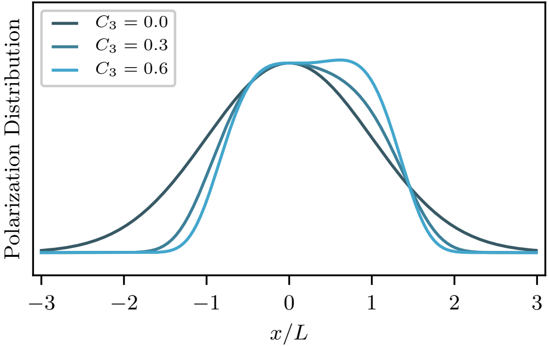
$$\frac{\pi e^2}{L^2 \hbar} C_2 = \int_0^\infty \frac{d\omega}{\omega} \mathrm{Re}\, \sigma(\omega).$$



I. Souza, T. Wilkens, R. M. Martin. PRB. 2000.

C_3

- C_3 is the *skew*.
- Quantifies imbalance between left and right shoulders of the distribution.



$C_1 \longleftrightarrow$ static response $\longleftrightarrow P$

$C_2 \longleftrightarrow$ linear response $\longleftrightarrow \sigma$

$C_3 \longleftrightarrow$ non-linear response? $\longleftrightarrow \boxed{?}$

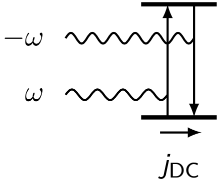
New Sum Rule

- For a two band system with time-reversal symmetry, diagonal responses obey

$$-\frac{\pi e^3}{2\hbar^2} C_3 = \int_0^\infty d\omega \; \sigma_{\text{shift}}(\omega).$$

- This is a new *nonlinear sum rule*.
- The **shift current** is the "solar panel response": how much DC current is generated from an applied electric field

$$\sigma_{\text{shift}}(\omega) = \text{Re}\, \sigma^{(2)}(0; \omega, -\omega).$$



- For two band systems, shift is related to SHG:

$$\sigma_{\text{SHG}}(\omega) = \text{Re}\, \sigma^{(2)}(-2\omega; \omega, \omega) = -2\sigma_{\text{shift}}(2\omega) + \sigma_{\text{shift}}(\omega).$$

N. Nagaosa, T. Morimoto. *Adv. Mater.* (2017).

Large C_3

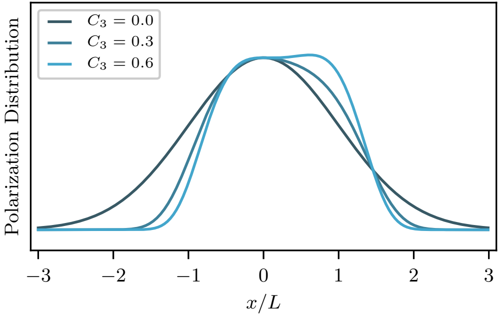
$\Updownarrow$

Large Shift Current

$\Updownarrow$

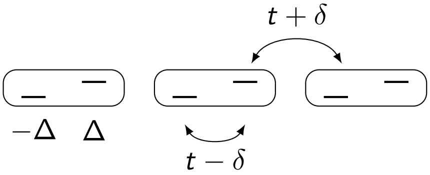
Large Second Harmonic Generation!

Real Space versus Reciprocal Space



$$\mathrm{Re}\,\sigma^{abc}_{\mathrm{SHG}}(-2\omega;\omega,\omega) = \frac{i\pi e^3}{2\hbar^2\omega^2}\sum_{m,n,p}\int\frac{d^dk}{(2\pi)^d}\Bigg[\left(v^a_{mn}w^{bc}_{nm} + 2v^a_{mn}\frac{v^b_{np}v^c_{pm} + v^c_{np}v^b_{pm}}{\omega_{mp}+\omega_{np}}\right) f_{mn}\delta_{2\omega-\omega_{nm}} + \left(w^{ab}_{mn}v^c_{nm} + w^{ac}_{mn}v^b_{nm}\right) f_{mn}\delta(\omega-\omega_{nm}) + v^a_{mn}\frac{v^b_{np}v^c_{pm}+v^c_{np}v^b_{pm}}{\omega_{pm}+\omega_{pn}}\Big(f_{mp}\delta(\omega-\omega_{pm}) - f_{np}\delta(\omega-\omega_{np})\Big)\Bigg]$$

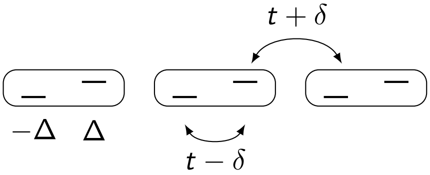
C_3 and TaAs



1. TaAs is quasi-1D.
2. TaAs is ferroelectric.
3. Rice-Mele is the cannonical 1D ferroelectric.

Therefore Rice-Mele is a good minimal model for TaAs.

C_3 and TaAs



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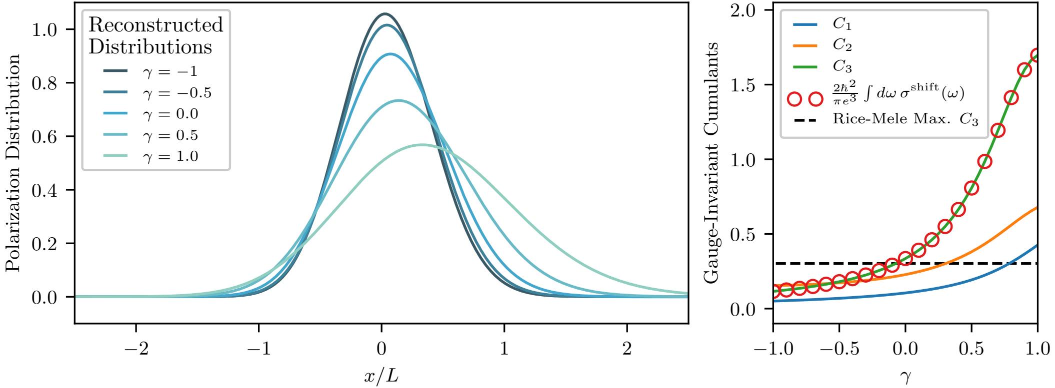
- There is a maximum C_3 in Rice-Mele:

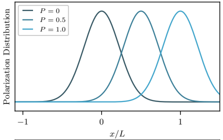
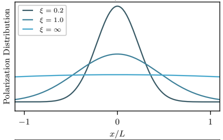
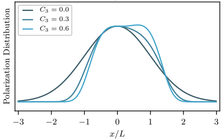
$$C_3 = \frac{e^3 a^2}{2\hbar^2} E_2(t/\Delta, \delta/\Delta) \leq 0.302 \frac{e^3 a^2}{2\hbar^2}$$

  where E_2 is the elliptic function of the second kind.
- The Rice-Mele parameters that describe TaAs well nearly reach this maximum.
- TaAs has essentially the largest SHG response for this class of materials.

Designing Large SHG



Summary

	Geometry	Visualization	Electric Response
C_1	Mean		Polarization
C_2	Variance/Localization		Conductivity
C_3	Skew		Second Harmonic