An invitation to Weyl semi-metals and their non-linear responses:

Outline

1) Non-linear optics
   * Zoology. / Symmetry.
   * Techniques. / Methods
   * Subtleties: A) disorder
     B) Gauges.
2) Examples:
   * Berry curvature dipole
   * Quantized injection current.

Refs: General response theory:

- Sipe & Shkrebtii PRB (2000)
- No time to read? Aversa, Sipe PRB, 52 14 636 (1995) until eq (11) then switch to appendix A of. de Juan et al. 1907.02537

1) Generalities.

What are we talking about?

$$j_i = \underbrace{\sigma^{(1)}_{ij}}_{\uparrow \text{ linear conductivity}} E_j + \underbrace{\sigma^{(2)}_{ijk}}_{\uparrow \text{ second-order conductivity}} E_j E_k + \dots$$

⚠ NOTE:
$$\left.\begin{array}{l} j \xrightarrow{I} -j \\ E \xrightarrow{I} -E \end{array}\right\} \text{ so } \sigma^{(2)} \text{ must be odd under inversion}$$

Now we want the response of the material in the long wavelength limit. $q \to 0$ and ω is finite.
i.e. we assume the wavelength is longer than sample size
good for optical range

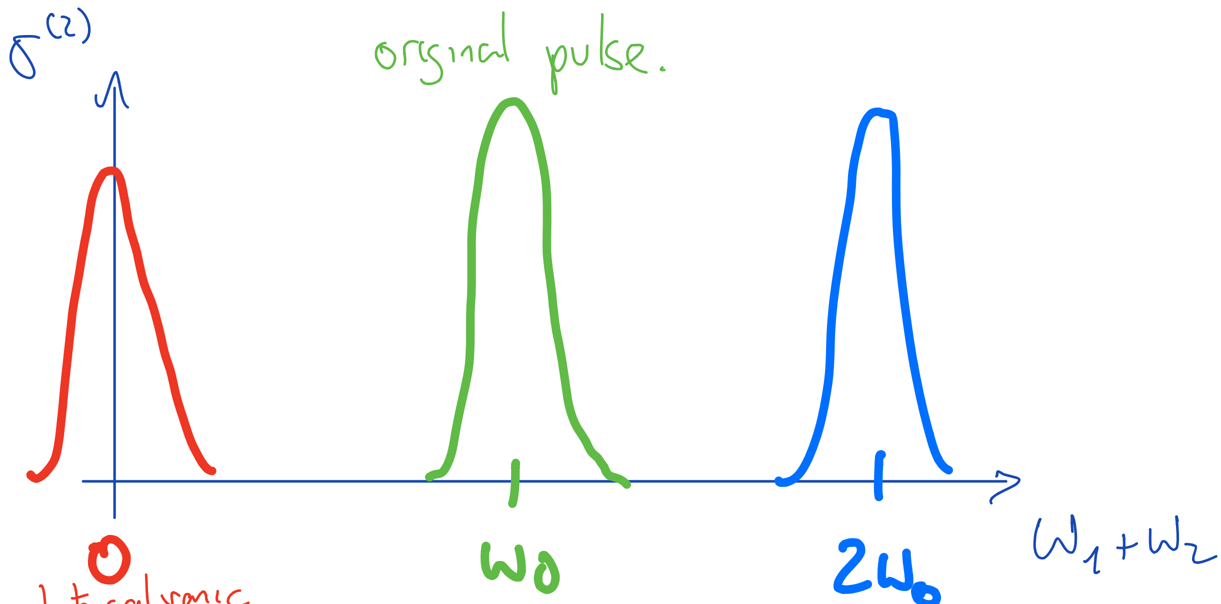
$$\boxed{\text{So } \quad \sigma^{(2)} \neq 0 \iff \text{inversion is broken}}$$

- same requirement as to have a WSM!
- also diagnosis for interacting systems b.t.w.

2) Hand wavy zoology of non-linear effects.

$$j(t) = \sigma^{(2)}(\omega_\alpha + \omega_\beta ; \omega_\alpha , \omega_\beta) E_{\omega_\alpha} E_{\omega_\beta} e^{i\omega_\Sigma t}$$

$\omega_\Sigma = \omega_\alpha + \omega_\beta$



photogalvanic effect.

a DC current from an oscillating field.

$j(0) \propto E_{\omega_0} E_{-\omega_0}$

second harmonic generation

→ laser with freq ω_0 gives response with $2\omega_0$

⚠ NOTE: There is also rectification "current"

= A constant polarization induced by light.

since $\vec{J} = \partial_t \vec{P}$

3) How do we calculate $\sigma^{(2)}_{ijk}(\omega_1+\omega_2\,;\,\omega_1,\omega_2)$?

- Semiclassical eqs of motion + Boltzmann eq. ~ finite τ
- Full quantum mechanical non-linear response ~ clean or disorder

Here we go into the subtleties:

WARNING 1:

finite disorder τ $\quad\neq\quad$ no τ from the start. $\left(\frac{1}{\tau}\to 0\right)$

$\omega\tau \to \infty$

(clean limit)

intrinsic contributions might cancel disorder induced but disorder independent contributions

references: old

Gerkin / Mednis / Belinicher ...

story

new

Pesin / Sodemann ...

WARNING 2: length gauge vs velocity gauge.

good refs: • F. Hipolito et al PRB 98, 205420 (2018) (length)

• D. J. Passos et al PRB 97, 235446 (2018) (velocity)

• G. B Ventura et al. PRB 96, 035431 (2017)

• Both give equivalent results but approximations made can lead to contradictory results.

$H_0 = \int d^3r \, \psi_r^\dagger H(r) \psi_r$

(Length or)
Position gauge: $H_p(t) = H_0 - q \vec{r} \cdot \vec{E}(t)$

Velocity gauge: $H_v(t) = H_0(\vec{p} - q\vec{A}(t))$

historically first → divergences → sum rules

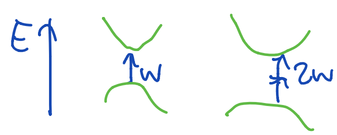
They are related by a unitary transformation:

$$U(t) = \exp\left[\frac{ie}{\hbar} \int d^d\vec{r} \, \vec{A}(t) \cdot \vec{r} \, \rho(\vec{r}) \right]$$

with $\rho(\vec{r}) = \psi^\dagger(\vec{r}) \psi(\vec{r})$ density operator.

$(U H_v U^\dagger + i\hbar(\partial_t U) U^\dagger = H_p)$

	PROS	CONS
Velocity gauge.	✓ can be formulated with Feynman rules Parker et al. PRB **99**, 045121 (2019) ✓ easier to implement numerically: $\langle n \vert v \vert m \rangle$ involved and not k derivatives of position operators. ✓ cleaner resonance structure (1-photon, 2-photon etc)	✗ spurious divergences if not careful enough. when $\omega \to 0$ (DC limit) ✗ Truncation over band sumations can give you spurious results.
Position gauge.	✓ Topological properties more transparent ✓ Difference between metals and insulators more transparent (de Juan et al. 1907.02537) ✓ easily connected to semiclassical responses ✓ Kramers-Kronigs rels allow numerics.	✗ Need to define position op in k space Blount (in 1960): $\langle mk \vert \vec{r} \vert nk' \rangle = +i\partial_k \delta_{kk'} \delta_{nm} + \delta_{kk'}\, i\langle n \vert \partial_k \vert m \rangle$ Berry conn $\downarrow$ A_{nm} $\langle mk \vert r_e \vert \ell k \rangle = (1-\delta_{m\ell}) A_{m\ell}\, \delta_{kk'}$ inter $\langle mk \vert r_i \vert \ell k \rangle = \delta_{m\ell}(\delta_{kk'} A_{m\ell} + i\partial_k \delta_{kk'})$ intra (only defined for $V \to \infty$) ←volume ✗ historically written with derivatives of position op



4) How do we calculate (Length gauge)

We want (1) $\langle \vec{J}(t) \rangle \overset{(*)}{=} \mathrm{Tr}\,(\rho(t)\vec{j}\,) = e\,\mathrm{Tr}(\rho\vec{v})$

e⁻ charge, velocity

(as any other expectation value)

density matrix

(*) $\langle A \rangle = \sum_i p_i \langle \psi_i | A | \psi_i \rangle = \sum_{\ell i} \langle \psi_i | \underbrace{p_i | \psi_\ell \rangle \langle \psi_\ell}_{\equiv \rho} | A | \psi_i \rangle \equiv \mathrm{Tr}[\rho A]$

we know $\vec{v} \overset{(**)}{=} \frac{1}{i\hbar} [H(t), \vec{r}\,]$ (2)

(**) from $\frac{d\langle \breve{A} \rangle}{dt} = \frac{1}{i\hbar} \langle [A, H] \rangle + \langle \frac{\partial \breve{A}}{\partial t} \rangle$

and $i\hbar \frac{\partial \rho}{\partial t} = [H, \rho]$

Combine (1) and (2) (Aversa Sipe)

+ Blount (position operator matrix elements in $H|m\rangle = \varepsilon_m|m\rangle$ basis)

The result is

$$\langle J^a \rangle = J_{intra} + \partial_t P_{inter}$$

(4) $$P^a_{inter} = e \int \frac{d k^d}{(2\pi)^d} \sum_{nm} r^a_{nm} \rho_{nm}(t)$$

(5) $$J^a_{intra} = e \int \frac{d k^d}{(2\pi)^d} \sum_{nm} D^a_{nm} \rho_{nm}(t)$$

'position operator'

$r^a_{nm} \equiv (1 - \delta_{nm}) A^a_{nm}$. (so $r_{nn} = 0$ hence "inter" label)

$A^a_{nm} \equiv i \langle n | \partial_k | m \rangle \equiv$ Berry connection

$\rho(t) = \sum_{nm} \rho_{nm}(t) |m\rangle\langle n|$; $\rho_{nm} \equiv \langle c^\dagger_n(k) c_m(k) \rangle$

$H|n\rangle = \varepsilon_n |n\rangle$

comes from $[H(t), r]\rho$

$$D^a_{nm} = v^a_{nn} \delta_{nm} - \frac{e}{\hbar} E^b(t) \left[r^b_{nm;a} + \delta_{nm} \epsilon^{dab} \Omega^d_n \right]$$

$v^a_{nn} \equiv \frac{\partial \varepsilon_n}{\partial k_a}$

generalized velocity; intraband vel.; interband vel.; anomalous vel.

$$r^b_{nm;a} \equiv \partial_{k_a} r^b_{nm} - i \left(A^a_{nn} - A^a_{mm} \right) r^b_{nm}$$

'generalized' derivative (under $|nk\rangle \to e^{i\theta^n_k} |nk\rangle$)

Now we want to get ρ_{nm} perturbatively.

The eq of motion of ρ is:

$$i\hbar\, \partial_t \rho = [\overset{H_0 + e\vec{r}\cdot\vec{E}}{H}, \rho] \quad (6)$$

$\downarrow H_0 \quad \searrow$ pert.

$$\Rightarrow \partial_t \rho_{nm} + i\,\varepsilon_{mn}\,\rho_{mn} = \frac{e E^b(t)}{\hbar}\Big[-\rho_{mn;b} + i\sum_p r^b_{mp}\rho_{pn} - \rho_{mp} r^b_{pn}\Big]$$

r_{intra} $\quad$ r_{inter}

when $E(t) = 0$ $\quad (7)\ \rho^{(0)}_{nm} = f(\varepsilon_n - \mu)\,\delta_{nm} \equiv f_n \delta_{nm}$.

i.e.: Filled states up to μ. with Fermi-Dirac distribution:

$$\rho = \sum_n f_n |n\rangle\langle n|$$

now $\quad E^b(t) = \sum_{\omega_\beta} E^b_\beta\, e^{-i\omega_\beta t}$

where β runs over different monochromatic components

e.g. Two frequency laser:

$$E^b(t) = E^b_1 e^{-i\omega_1 t} + E^{b*}_1 e^{i\omega_1 t} + E^b_2 e^{-i\omega_2 t} + E^{b*}_2 e^{i\omega_2 t}$$

Linear response

Now combine (7) and (6) and solve for $\rho_{nm} = \rho^0_{nm} + \rho^{(1)}_{nm} + \dots$

$$\rho^{(1)}_{nm}(t) = -\frac{e}{\hbar}\sum_\beta \mathcal{E}^{b}_\beta e^{-i\omega_\beta t}\left[\frac{f_{nm} r^b_{mn}}{\omega_\beta - \varepsilon_{mn}} + i\,\frac{f_{n,b}\,\delta_{mn}}{\omega_\beta}\right]$$

$\varepsilon_{mn} \equiv \varepsilon_m - \varepsilon_n$

Then we plug it in (4) and (5) to obtain the linear response: Choose $\mathcal{E}^b(t) = (e^{-i\omega t} + e^{i\omega t})\mathcal{E}^b$

$$J(t) = J_{intra}(t) + \partial_t P_{inter} \equiv \sum_{\omega_\beta = \pm\omega} \sigma^{ab}(\omega_\beta)\mathcal{E}^b_\beta e^{-i\omega_\beta t}$$

$$\sigma^{ab}(\omega) = \frac{e^2}{\hbar}\int_k \sum_{nm}\left[i\omega(f_n - f_m)\frac{r^a_{nm} r^b_{mn}}{(\omega - \varepsilon_{mn})} - \delta_{mn} f_n\left(\frac{\omega_{n,ba}}{i\omega} + \epsilon^{abc}\Omega_c\right)\right]$$

2nd-order resp.

Then we do the same and get $\rho^{(2)}_{nm}$.

Then we plug that back into (4) (5) to obtain for the non-linear response:

$$\partial_t P^a_{inter}(t) = -i\omega_\Sigma \chi^{abc(2)}_{inter}(\omega_\beta, \omega_\gamma)\mathcal{E}^b_\beta \mathcal{E}^c_\gamma e^{-i\omega_\Sigma t}$$

$$J^b_{intra}(t) = \chi^{abc(2)}_{intra}(\omega_\beta, \omega_\gamma)\mathcal{E}^b_\beta \mathcal{E}^c_\gamma e^{-i\omega_\Sigma t}$$

$$\omega_\Sigma \equiv \omega_\gamma + \omega_\beta$$

Focus on DC response: $\omega_\Sigma \simeq 0$ so $\partial_t P_{inter} \propto \omega_\Sigma \ll 1$

Very complicated expression (see de Juan)

but simplifies a lot focusing on $\omega_\Sigma \simeq 0$

$\omega_1 = \omega + \Delta\omega/2$

$\omega_2 = \omega - \Delta\omega/2$

$\Delta\omega \ll 1$. + Time reversal symmetry

monochromatic limit

not really important but simplifies

$$J(t) = 4\left[\frac{\sin(\Delta\omega t)}{\Delta\omega}\beta^{ab}(\omega) + \cos(\Delta\omega t)\gamma^{ab}(\omega)\right]\underbrace{(\vec{E}\times\vec{E}^*)^b}_{c}$$

$$+ 2\cos(\Delta\omega t)\,\sigma^{abc}(\omega)\left(E^b E^{c*} + E^c E^{b*}\right)$$

$\omega_\alpha = \omega - \frac{\Delta\omega}{2}$ $\omega_\beta = -\omega - \frac{\Delta\omega}{2}$, $\omega_\alpha = \omega + \frac{\Delta\omega}{2}$

$\omega_\beta = -\omega + \frac{\Delta\omega}{2}$

Things we notice:

linear polarization

$\sigma^{abc}(\omega) \equiv$ shift current. if large, this is good for photocells!

$$\underset{\in\mathbb{R}}{\sigma^{abc}(\omega)} = \frac{\pi C}{2}\int_k \sum_{n>m} f_{nm}\mathrm{Im}[r^b_{mn} r^c_{nm;a}]\delta(\varepsilon_{mn} - \omega) \quad (3)$$

* we know how to make it large ~ longer range hopp.

* can be shown to be related to polarization shift of photoexcited e^-.

Circular polarization. e.g. $|E|(1, \pm i, 0)$

$\gamma^{ab} \Rightarrow$ a Fermi surface integral

only non-zero for metals e.g. Weyl.

but non-topological.

+ Berry curvature dipole. (Sodeman / Fu, Moore / Orenstein)

(a semiclassical term) + extra new piece.

very dispersive with divergences.

$$\gamma_1^{ab}(\omega) = \frac{iC}{2} \int_k \sum_{n>m} \frac{\omega f_{nm,a}}{\varepsilon_{nm}^2 - \omega^2} \mathrm{Im}[\epsilon^{bcd} r_{nm}^d r_{mn}^c] \qquad (4)$$

$$\gamma_2^{ab}(\omega) = -\frac{iC}{2\omega} \int_k \sum_n [(f_{n,a}\Omega_n^b - \delta^{ab} f_{n,c}\Omega_n^c)] \qquad (5)$$

measures separation btw regions of $\pm\Omega$.

$\hookrightarrow$ Fu: $\frac{\tau}{1+\omega\tau} \int_{k} f_0 \, \partial\Omega$

$\omega\tau \to \infty$ clean limit

$\beta^{ab}(\omega)$ is really a current that grows in time for $\Delta\omega \ll 1$

$$\beta^{ab}(\omega) = \frac{i\pi C}{4} \int_k \sum_{n>m} f_{nm} \Delta_{mn}^a \mathrm{Im}[r_{nm}^d r_{mn}^c] \epsilon^{bcd} \delta(\varepsilon_{mn} - \omega)$$

$v_{nn}^a - v_{mm}^b \equiv \partial \varepsilon_{nm}$

normal to the $\varepsilon_{nm} = \omega$ surface

$$\frac{\partial_{k_a} \varepsilon_{nm}}{\varepsilon_{nm}} = \hat{n}_a^k \cdot \Omega^a$$

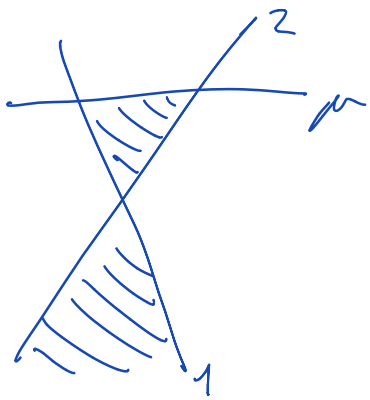
Lets focus on $\beta^{ab}(\omega)$: for $\Delta\omega \to 0$ we see:

$$\frac{dJ(t)}{dt} = \beta^{ab}(\omega)\left[\vec{E} \times \vec{E}^*\right]^b$$

$\equiv$ injection current.

Why is it special? Only special for Weyls: Lets calculate it:

We need:



only $m \to n$
$1 \to 2$

$f_n = \Theta(\mu - \epsilon_n)$ so:

$f_1 = 1$ for all k's. $= \Theta(v_F|k|)$

$f_2 = \Theta(\mu - \epsilon_2)$

Sum rule:

$$i \sum_{n \neq m} \epsilon_{cab} r_{nm}^b r_{mn}^a = \Omega_n^c$$

$$\Rightarrow \quad i\,\epsilon_{cab}\, r_{12}^b r_{21}^c = \Omega_1^c = \frac{1}{2}\frac{k^c}{|k|^3}$$

$$\Rightarrow \Delta_{21}^a = \partial_{k_a}(v_F|k|) - \partial_{k_a}(-v_F|k|) = -2\, k^a/|k|\, v_F = -2\hat{k}^a v_F$$

$$\Rightarrow \delta(\epsilon_{12} - \hbar\omega) = \delta(|k| - k(\omega))/2v_F \qquad k(\omega) = \frac{\omega}{2v_F}$$

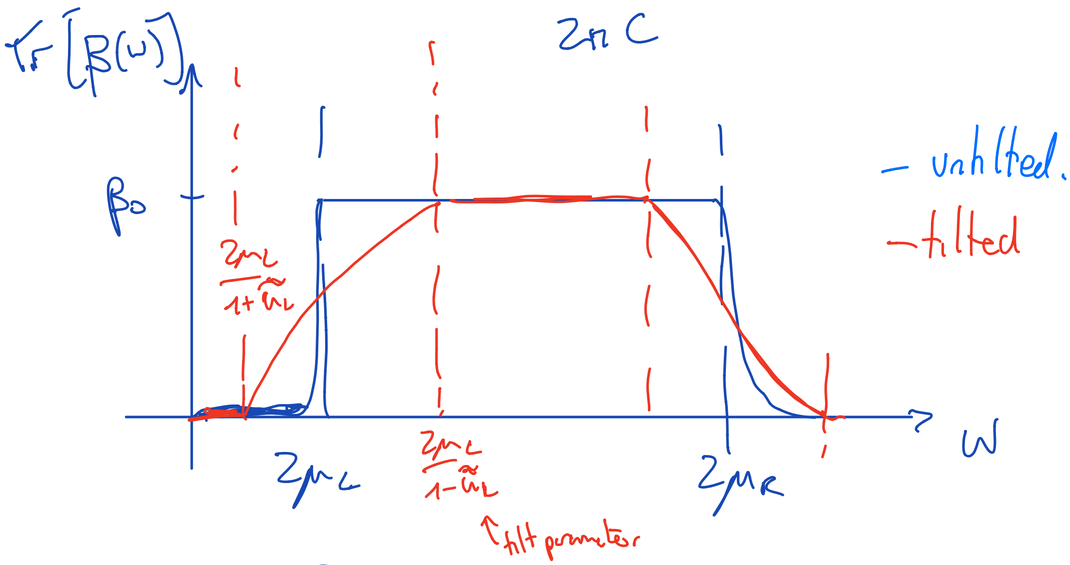
And calculate the Trace of β.

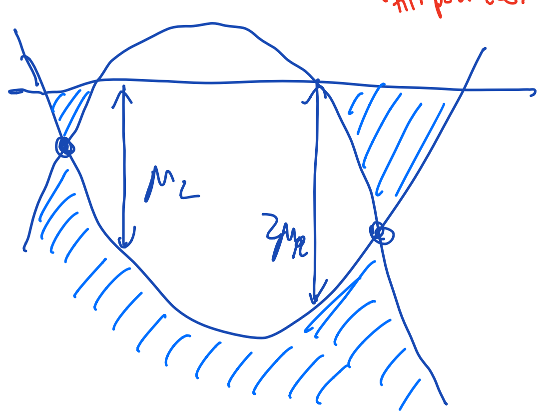
$$\mathrm{Tr}[\beta] = \frac{ie^3\pi}{\hbar^2}\int \frac{d\Omega_{\text{solid}}}{(2\pi)^3}\int k^2 dk\, \Theta(\underbrace{2v_F|k|}_{\omega}-2\mu)\,\frac{2v_F k^i}{|k|}\,\frac{1}{2}\frac{k^i}{|k|^3}\,\frac{\delta(k-k(\omega))}{2v_F}$$

$$= \frac{ie^3\pi}{\hbar^2}\Theta(\omega-2\mu) \equiv i\beta_0\,\Theta(\omega-2\mu)$$

This is general for two band models: (go back to formula)

$$\mathrm{Tr}[\beta] = \frac{ie^3}{2\hbar^2}\underbrace{\oint d\vec{S}_k\cdot\vec{\Omega}_k}_{2\pi C} = C\,\frac{i\pi e^3}{\hbar^2}$$



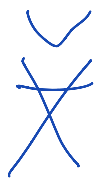


needs mirror breaking!

chiral crystals!

Some comments:

- 2 bands quantization is exact. but



$\Delta\epsilon_{23}$ gives corrections:

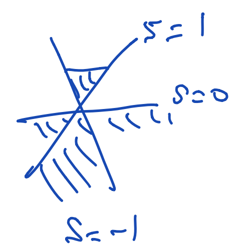
$$\mathrm{Tr}[\beta] = i\beta_0 + \mathcal{O}\left(\frac{\Delta\epsilon_{23}}{w^2}\right)$$

- Sometimes nature is kind and matrix elements can vanish. making corrections zero.

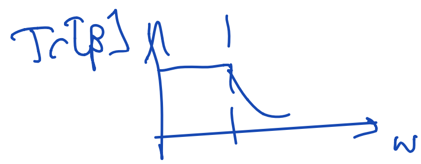
↳ multifold fermions:

$$H = v_F \vec{k}\cdot\vec{S}$$

↑ spin matrix



- Something similar measured in RhSi (Berkeley)



For you:

- interacting version?
- protected quantized non-linear?
- disorder corrections?

Cheat sheet for non-linear (see appendix in de Juan et al. 1907.02537)

- $R_{nm}^{j} = \epsilon_{jkl} \, r_{nm}^{k} r_{mn}^{l}$

- $\Omega_n^c = i \sum_{n \neq m} R_{nm}^{c} = i \sum_{n\neq m} \epsilon_{cab} \, r_{nm}^{a} r_{mn}^{b}$

- $\epsilon_{ijk} \epsilon_{ljk} = 2\delta_i^l \qquad \epsilon^{abc}\epsilon_{emc} = \delta_e^a \delta_m^b - \delta_m^a \delta_e^b$

- $r_{nm}^{b} r_{mn}^{c} = \left(r_{nm}^{c} r_{mn}^{b}\right)^{*}$ $\begin{cases} \text{Re is symmetric} \\ \text{Im is antisymmetric} \end{cases}$

- $F_\pm = \dfrac{1}{\omega_{mn}-\omega} \pm \dfrac{1}{\omega_{mn}+\omega} + i\pi\left[-\delta(\omega_{mn}-\omega) \pm \delta(\omega_{mn}+\omega)\right]$

- $F_\pm^{*}(\omega_{mn}, -\omega) = \pm F_\pm^{*}(\omega_{mn}, \omega)$

- An antisymmetric tensor $A_{abc} = \frac{1}{2}\left(T_{abc} - T_{acb}\right) = \epsilon_{bcd} B^{ad}$
  and $B_{ad} = \frac{1}{2} \epsilon_{dbc} A^{abc}$

- $f_{n,a} = \partial_{ka} \Theta(\mu - \epsilon_k) = v^a \partial_\epsilon \Theta(\mu - \epsilon) = -v^a \delta(\mu - \epsilon)$