Trion in time-resolved ARPES

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Theory of TR-ARPES

The most generic theory requires Keldysh formalism but let's spare ourselves the burden. . .

Ingredients of our model of ARPES

- Electric dipole interaction only
- Sudden approximation
- *Separation between pump and probe*: system *not* driven when probed; pump prepares an initial state and nothing more
- Fermi's golden rule in probing

Theory of TR-ARPES

Main result Output intensity $I_{\mathbf{k}}(t) \propto \sum_c P^c_{\mathbf{k}}(t)$,

$$P^c_{\mathbf{k}}(t) = \sum_{n,\mathbf{k}'} \rho_n |M^{fc}_{\mathbf{k}\mathbf{k}'}|^2 \int_{t_0}^t \mathrm{d}t_1 \int_{t_0}^t \mathrm{d}t_2 \mathrm{e}^{-\mathrm{i}(E_n - \omega)(t_1 - t_2)} \langle \Psi_n(t_0) | c^\dagger_{\mathbf{k}'} U(t_2, t_1) c_{\mathbf{k}'} | \Psi_n(t_0) \rangle s(t_1) s(t_2).$$

1. ρ_n: distribution of the final state of pumping (initial state of probing).
2. Probe field is $\mathbf{E} = s(t)\mathbf{E}_0 \mathrm{e}^{-\mathrm{i}\omega_0 t} + \text{c.c.}$, and the transition matrix is $M^{fc}_{\mathbf{k}\mathbf{k}'} = \langle f\mathbf{k} | {-\mathbf{d}} \cdot \mathbf{E}_0 | c\mathbf{k}' \rangle$; here c is the band that is interacting with light, f is the out-going state.
3. The $\mathrm{e}^{-\mathrm{i}(E_n - \omega)(t_1 - t_2)}$ factor gives half of energy conservation condition; ω is driving frequency ω_0 shifted by work function.
4. $\langle \Psi_n | \cdots | \Psi_n \rangle$: electron Green function with excited state background $|\Psi_n\rangle$; it gives the structure of $|\Psi_n\rangle$ in electron basis, and the second half of energy conservation condition (energy after one electron being kicked out);
5. $s(t_1)s(t_2)$: shape of probe pulse; broadening $\delta(E_n - \omega - E_{\text{after}})$

Successful example: exciton

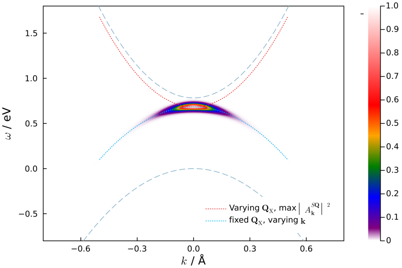
- Below we work with 2D material $\Rightarrow \mathbf{k}_{\parallel}^{\text{out}} = \mathbf{k}'$; we refer to $\mathbf{k}_{\parallel}^{\text{out}}$ as $\mathbf{k}$
- Only consider valence band top and conduction band valley: parabolic bands

ARPES response of an exciton (besides valence band electron)

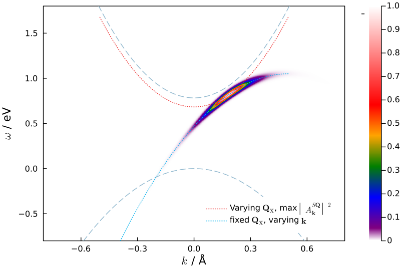
$$P(t)=\sum_{S,\mathbf{Q}} \rho_{S\mathbf{Q}} \left|A_{\mathbf{k}'}^{S\mathbf{Q}}\right|^2 \left|M_{\mathbf{k}\mathbf{k}'}^{fc}\right|^2 \int_{t_0}^{t} \mathrm{d}t_1 \int_{t_0}^{t} \mathrm{d}t_2 \, \mathrm{e}^{-\mathrm{i}(E_{S\mathbf{Q}}+\epsilon_{v\mathbf{k}'-\mathbf{Q}}-\omega)(t_1-t_2)} s(t_1) s(t_2).$$

- Detection of $\left|A_{\mathbf{k}}^{S\mathbf{Q}}\right|^2$
- $\mathbf{Q} = 0 \Rightarrow$ dispersion relation $\omega = \epsilon_{v\mathbf{k}}$ (energy conservation)
- $\mathbf{Q}$ obeys $\mathrm{e}^{-\beta E_{\mathbf{Q}}}$ distribution: dispersion relation is $\omega = E_{\mathrm{g}} + E_{\mathrm{B}} + \frac{\mathbf{k}^2}{2m_{\mathrm{e}}}$ (energy conservation; $\left|A_{\mathbf{k}}^{S\mathbf{Q}}\right|^2$ reaches peak when $\mathbf{v}_{\mathrm{e}} = \mathbf{v}_{\mathrm{h}}$)

Successful example: exciton



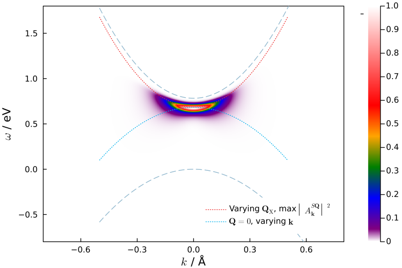
Single exciton, $\mathbf{Q} = 0.0\,\text{Å}^{-1}$



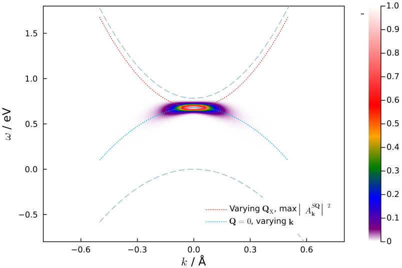
Single exciton, $\mathbf{Q} = 0.5\,\text{Å}^{-1}$

- The center of the signature is on a replica of the conduction band
- The shape of the signature is a replica of the valence band

Successful example: exciton



Thermalized exciton, $\beta = 10\,\mathrm{eV}$



Thermalized exciton, $\beta = 100\,\mathrm{eV}$

- The center of the signature is on a replica of the conduction band
- The shape of the signature is a replica of the valence band
- When excitons are hot, the dispersion relation is the conduction band; when they are cold, the dispersion relation is the valence band

Trion: some theoretical issues

In the current results:

- Two holes, one electron (what likely happens in ARPES)
- *Two holes on one band* – to simply analysis
- *No scattering after the electron is kicked out* – realistic or not?

Trion Hamiltonian By some analysis:

$$
\begin{aligned}
H_{\mathrm{T}} &= \frac{\mathbf{k}_{\mathrm{e}}^2}{2m_{\mathrm{e}}} + \frac{\mathbf{k}_{\mathrm{h1}}^2}{2m_{\mathrm{h}}} + \frac{\mathbf{k}_{\mathrm{h2}}^2}{2m_{\mathrm{h}}} + V(\mathbf{r}_{\mathrm{h1}} - \mathbf{r}_{\mathrm{h2}}) - V(\mathbf{r}_{\mathrm{e}} - \mathbf{r}_{\mathrm{h1}}) - V(\mathbf{r}_{\mathrm{e}} - \mathbf{r}_{\mathrm{h2}}) \\
&= \underbrace{H_{\mathrm{X}}(\mathbf{r}_1) + H_{\mathrm{X}}(\mathbf{r}_2) - \frac{\hbar^2 \nabla^2}{2\mu} + V(\mathbf{r}_1 - \mathbf{r}_2) - E_{\mathrm{g}}}_{\text{causing } E_{\mathrm{B}}} + \frac{\mathbf{P}_{\mathrm{T}}^2}{2m_{\mathrm{T}}} + E_{\mathrm{g}},
\end{aligned}
\tag{1}
$$

where

$$
m_{\mathrm{T}} = \underbrace{2m_{\mathrm{h}}}_{>0} + m_{\mathrm{e}}, \quad \mathbf{k}_{\mathrm{h1}} = \mathbf{k}_1 + \frac{m_{\mathrm{h}}}{m_{\mathrm{T}}}\mathbf{P}_{\mathrm{T}}, \quad \mathbf{k}_{\mathrm{h2}} = \mathbf{k}_2 + \frac{m_{\mathrm{h}}}{m_{\mathrm{T}}}\mathbf{P}_{\mathrm{T}}, \quad \mathbf{k}_{\mathrm{e}} = \frac{m_{\mathrm{e}}}{m_{\mathrm{T}}}\mathbf{P}_{\mathrm{T}} - \mathbf{k}_1 - \mathbf{k}_2. \tag{2}
$$

That's to say: a trion, when the two holes of it are of the same species, can be seen as the bound state of two excitons.

Trion wave function Thus in this specific case:

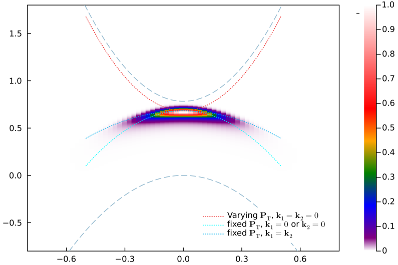
$$\phi_{\mathrm{T}}(\mathbf{r}_{\mathrm{e}}, \mathbf{r}_{\mathrm{h}1}, \mathbf{r}_{\mathrm{h}2}) = \sum_{S_1, S_2} \phi_{\mathrm{X}}^{S}(\mathbf{r}_1) \phi_{\mathrm{X}}^{S}(\mathbf{r}_2) \, \mathrm{e}^{\mathrm{i} \, \mathbf{P}_{\mathrm{T}} \cdot \mathbf{R}}, \tag{3}$$

where $\mathbf{r}_{1,2}$ are defined above.

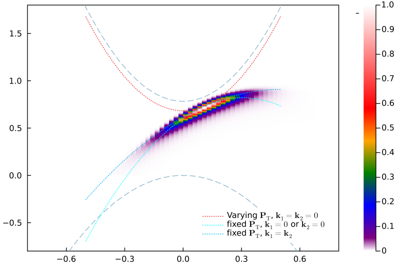
In the tentative plots below We ignore spin and choose $S_1 = S_2 = 1\mathrm{s}$. The ARPES response (here ϵ_v are *negative*):

$$\begin{aligned} P_{\mathbf{k}}(t) = {} & \sum_{\mathbf{P}_{\mathrm{T}}} \rho_{\mathbf{P}_{\mathrm{T}}} \sum_{\mathbf{k}_{\mathrm{h}1}, \mathbf{k}_{\mathrm{h}2}} \sum_{S_1, S_2} \left| M_{\mathbf{k}\mathbf{k}_{\mathrm{e}}}^{fc} \right|^2 \left| A_{S_1 S_2} \right|^2 \left| \phi_{S_1}(\mathbf{k}_1) \right|^2 \left| \phi_{S_2}(\mathbf{k}_2) \right|^2 \\ & \times \int_{t_0}^{t} \mathrm{d}t_1 \int_{t_0}^{t} \mathrm{d}t_2 \, \mathrm{e}^{-\mathrm{i}(E_{S\mathbf{P}_{\mathrm{T}}} + \epsilon_{v\mathbf{k}_{\mathrm{h}1}} + \epsilon_{v\mathbf{k}_{\mathrm{h}2}} - \omega)(t_1 - t_2)} \, s(t_1) s(t_2) \end{aligned} \tag{4}$$

Trion: tentative results

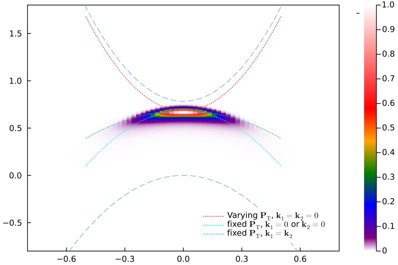


Single trion, $\mathbf{P} = 0.0\,\text{Å}^{-1}$



Single trion, $\mathbf{P} = 0.5\,\text{Å}^{-1}$

- The center of the signature is still sticked to a replica of the conduction band (moved downwards by E_B): the maximum of a signature is reached when $\mathbf{k}_1 = \mathbf{k}_2 = 0$ where $E_\mathrm{T} = \epsilon_v - E_\mathrm{B}$



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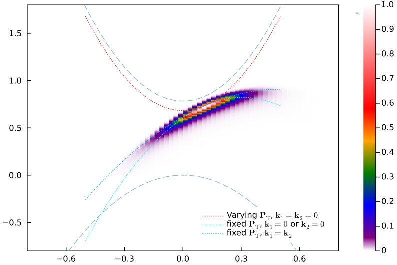
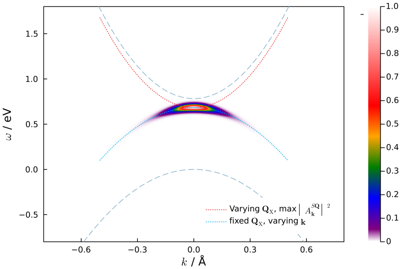


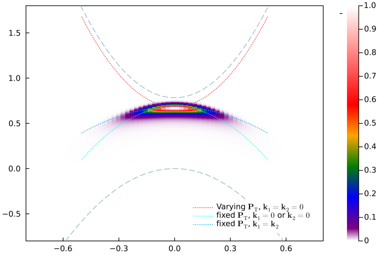
Single trion, $\mathbf{P} = 0.5\,\text{Å}^{-1}$

- The analytic form of the shape of a single-trion signature is still not clear; I tried to set one of $\mathbf{k}_{1,2}$ to zero, or $\mathbf{k}_1 = \mathbf{k}_2$; both of them seem to work, but not that well; the dispersion relation given by the first (but not the second) condition is also a replica of ϵ_v.

Trion: tentative results

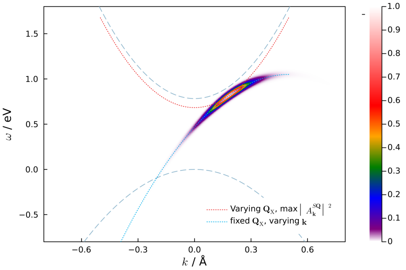


Exciton, $\mathbf{Q} = 0.0\,\text{Å}^{-1}$

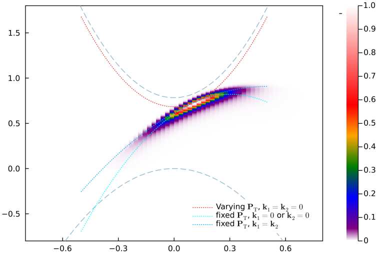


Trion, $\mathbf{P} = 0.0\,\text{Å}^{-1}$

Trion: tentative results



Exciton, $\mathbf{Q} = 0.5\,\text{Å}^{-1}$



Trion, $\mathbf{P} = 0.5\,\text{Å}^{-1}$

Conclusion

- Trion signature on ARPES spectrum looks similar to that of exciton
- The finite-T induced "upper" dispersion of the two is the same
- But the shape and spread of a single trion mode is still different from those of an exciton mode, even with close E_{B}