

Trion in time-resolved ARPES

Jinyuan Wu

November 8, 2023

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 $I_{\mathbf{k}}(t) \propto \sum_c P_{\mathbf{k}}^c(t)$,

$$P_{\mathbf{k}}^c(t) = \sum_{n,k'} \rho_n |M_{\mathbf{k}\mathbf{k}'}^{fc}|^2 \int_{t_0}^t dt_1 \int_{t_0}^t dt_2 e^{-i(E_n - \omega)(t_1 - t_2)} \langle \Psi_n(t_0) | c_{\mathbf{k}'}^\dagger U(t_2, t_1) c_{\mathbf{k}} | \Psi_n(t_0) \rangle s(t_1) s(t_2).$$

- ① ρ_n : distribution of the final state of pumping (initial state of probing).
- ② Probe field is $\mathbf{E} = s(t) \mathbf{E}_0 e^{-i\omega_0 t} + \text{c.c.}$, and the transition matrix is $M_{\mathbf{k}\mathbf{k}'}^{fc} = \langle f\mathbf{k} | -\mathbf{d} \cdot \mathbf{E}_0 | c\mathbf{k}' \rangle$; here c is the band that is interacting with light, f is the out-going state.
- ③ The $e^{-i(E_n - \omega)(t_1 - t_2)}$ 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

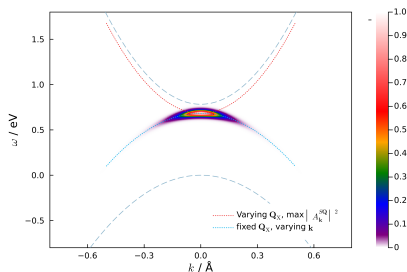
- Below we work with 2D material $\Rightarrow \mathbf{k}_{\parallel}^{\text{out}} = \mathbf{k}'$; we refer to $\mathbf{k}_{\parallel}^{\text{out}}$ as \mathbf{k}
- Only consider valence band top and conduction band valley: parabolic bands

ARPES response of an exciton (besides valence band electron)

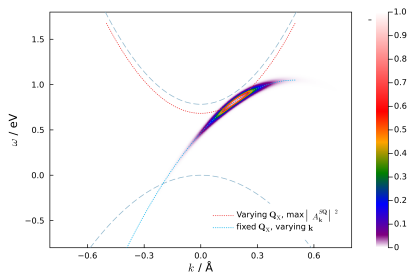
$$P(t) = \sum_{S, \mathbf{Q}} \rho_{S\mathbf{Q}} |A_{\mathbf{k}'}^{S\mathbf{Q}}|^2 |M_{\mathbf{k}\mathbf{k}'}^{fc}|^2 \int_{t_0}^t dt_1 \int_{t_0}^t dt_2 e^{-i(E_{S\mathbf{Q}} + \epsilon_{v\mathbf{k}'} - \mathbf{Q} - \omega)(t_1 - t_2)} s(t_1) s(t_2).$$

- Detection of $|A_{\mathbf{k}}^{S\mathbf{Q}}|^2$
- $\mathbf{Q} = 0 \Rightarrow$ dispersion relation $\omega = \epsilon_{v\mathbf{k}}$ (energy conservation)
- \mathbf{Q} obeys $e^{-\beta E_{\mathbf{Q}}}$ distribution: dispersion relation is $\omega = E_g + E_B + \frac{\mathbf{k}^2}{2m_e}$ (energy conservation; $|A_{\mathbf{k}}^{S\mathbf{Q}}|^2$ reaches peak when $\mathbf{v}_e = \mathbf{v}_h$)

Successful example: exciton



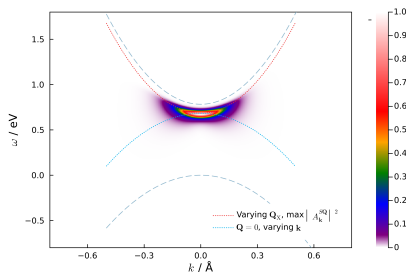
Single exciton, $Q = 0.0 \text{ \AA}^{-1}$



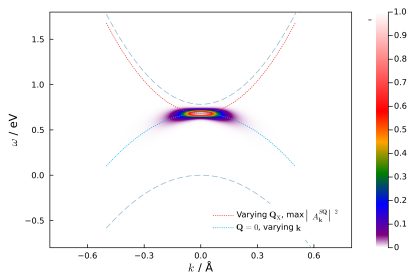
Single exciton, $Q = 0.5 \text{ \AA}^{-1}$

- 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, $\beta = 10$ eV



Thermalized exciton, $\beta = 100$ 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:

$$\begin{aligned} H_T &= \frac{\mathbf{k}_e^2}{2m_e} + \frac{\mathbf{k}_{h1}^2}{2m_h} + \frac{\mathbf{k}_{h2}^2}{2m_h} + V(\mathbf{r}_{h1} - \mathbf{r}_{h2}) - V(\mathbf{r}_e - \mathbf{r}_{h1}) - V(\mathbf{r}_e - \mathbf{r}_{h2}) \\ &= \underbrace{H_X(\mathbf{r}_1) + H_X(\mathbf{r}_2) - \frac{\hbar^2 \nabla^2}{2\mu} + V(\mathbf{r}_1 - \mathbf{r}_2) - E_g}_{\text{causing } E_B} + \frac{\mathbf{P}_T^2}{2m_T} + E_g, \end{aligned} \quad (1)$$

where

$$m_T = \underbrace{2m_h}_{>0} + m_e, \quad \mathbf{k}_{h1} = \mathbf{k}_1 + \frac{m_h}{m_T} \mathbf{P}_T, \quad \mathbf{k}_{h2} = \mathbf{k}_2 + \frac{m_h}{m_T} \mathbf{P}_T, \quad \mathbf{k}_e = \frac{m_e}{m_T} \mathbf{P}_T - \mathbf{k}_1 - \mathbf{k}_2. \quad (2)$$

Trion: some theoretical issues

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:

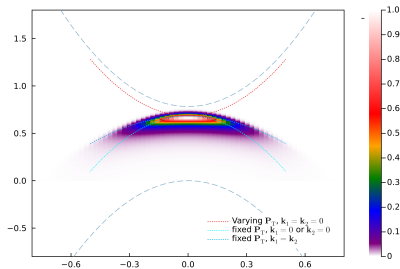
$$\phi_T(\mathbf{r}_e, \mathbf{r}_{h1}, \mathbf{r}_{h2}) = \sum_{S_1, S_2} \phi_X^S(\mathbf{r}_1) \phi_X^S(\mathbf{r}_2) e^{i\mathbf{P}_T \cdot \mathbf{R}}, \quad (3)$$

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

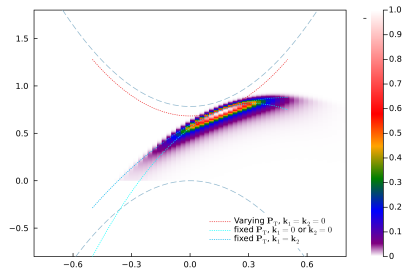
In the tentative plots below We ignore spin and choose $S_1 = S_2 = 1s$. The ARPES response (here ϵ_v are *negative*):

$$P_{\mathbf{k}}(t) = \sum_{\mathbf{P}_T} \rho_{\mathbf{P}_T} \sum_{\mathbf{k}_{h1}, \mathbf{k}_{h2}} \sum_{S_1, S_2} |M_{\mathbf{k}\mathbf{k}_e}^{fc}|^2 |A_{S_1 S_2}|^2 |\phi_{S_1}(\mathbf{k}_1)|^2 |\phi_{S_2}(\mathbf{k}_2)|^2 \quad (4)$$
$$\times \int_{t_0}^t dt_1 \int_{t_0}^t dt_2 e^{-i(E_{S\mathbf{P}_T} + \epsilon_{v\mathbf{k}_{h1}} + \epsilon_{v\mathbf{k}_{h2}} - \omega)(t_1 - t_2)} s(t_1) s(t_2)$$

Trion: tentative results



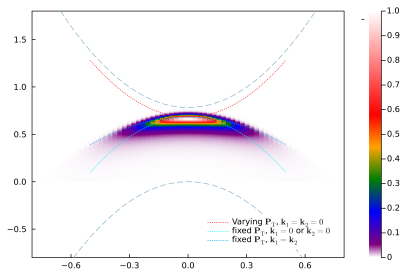
Single trion, $P = 0.0 \text{ \AA}^{-1}$



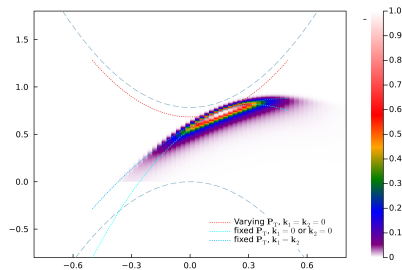
Single trion, $P = 0.5 \text{ \AA}^{-1}$

- The center of the signature is still stuck 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 $E_T = \epsilon_v - E_B$

Trion: tentative results



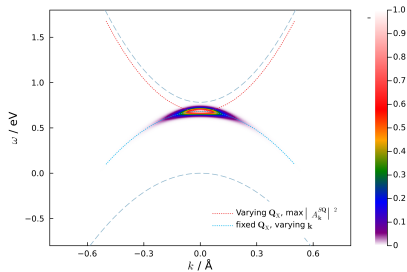
Single trion, $P = 0.0 \text{ \AA}^{-1}$



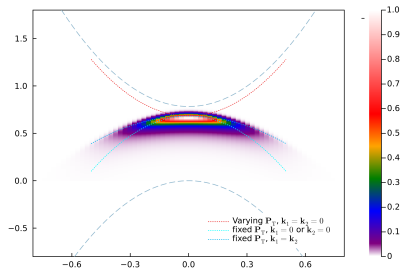
Single trion, $P = 0.5 \text{ \AA}^{-1}$

- 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

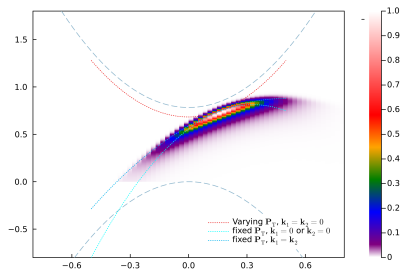
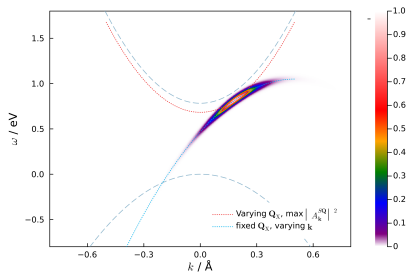


Exciton, $\mathbf{Q} = 0.0 \text{ \AA}^{-1}$



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

Trion: tentative results



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