Preliminary results on trion signature in time-resolved ARPES

When the temporal separation between the pump and the probe is long enough, the effect of the pump is essentially preparing an initial state with various excitations, which are then detected by the probe. The probe drives the electron part(s) of an excitation out the material, which then contributes to the total ARPES intensity; since the electrons in the excitations always reside on conduction bands, they leave signatures in the resulting ARPES spectrum that are absent in conventional ARPES. The probability to detect an electron with momentum $\mathbf{k}$ from an excitation $|\Psi_n\rangle$ at time t is given as [1]

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

(1)

where t_0 is the time the probe starts, ω is the energy of the out-coming electron plus the work function minus the photon energy, E_n is the energy of $|\Psi_n(t)\rangle$, c is a band index in the material, $c_{\mathbf{k}'}$ refers to the annihilation operator of that mode, $M_{\mathbf{kk}'}^{fc}$ is the dipole transition matrix between the $(c, \mathbf{k}')$ band electron state in the material and the outgoing final state $(f, \mathbf{k})$, $U(t_2, t_1)$ is the many-body time evolution operator in the material, and $s(t)$ is the shape of the probe pulse. The time evolution operator carries information regarding the oscillation of the state of the system after one electron is kicked out, leading to a broadened form of energy conservation after the integrations over t_1 and t_2 are finished that determines the shape of the ARPES signature. It is possible that the final state of pumping is a mixed state of several excitations, and in this case (1) needs to be averaged over the probabilistic distribution of the excitations, and the excitation signatures are overlaid to each other.

In the case of two-dimensional materials, $\mathbf{k}_{\parallel} = \mathbf{k}'$. Within the framework of the two-band model, it has been shown [1] that the signature of an exciton has the dispersion relation of

$$\omega = E_{S\mathbf{Q}} + \epsilon_{v\mathbf{k}_{\parallel}-\mathbf{Q}}, \tag{2}$$

where $E_{S\mathbf{Q}}$ is the single exciton energy, i.e. band gap plus the negative binding energy, and $\epsilon_{v\mathbf{k}} < 0$ is the valence band energy from the Fermi energy. In the case of a positive trion, there is no single, definite dispersion relation that determines the shape of the ARPES signature; assuming that after one electron is driven out, scatterings between the two remaining holes are negligible, the dispersion of the trion ARPES signature is determined by the following family of curves

$$\omega = E_{S\mathbf{P}} + \epsilon_{v\mathbf{k}_{\mathrm{h}1}} + \epsilon_{v\mathbf{k}_{\mathrm{h}2}}, \quad \mathbf{k}_{\parallel} - \mathbf{k}_{\mathrm{h}1} - \mathbf{k}_{\mathrm{h}2} = \mathbf{P}. \tag{3}$$

In Fig. 1, it could be observed that in the zero-temperature case, even when a trion mode and an exciton mode with the same energy, they still result in different ARPES signatures: the slopes and curvatures of the ARPES signature are different, and when the total momentum of the excitation is given, the position shifts of the signature on the ARPES heatmap are also different.

References

[1] Avinash Rustagi and Alexander F Kemper. "Photoemission signature of excitons". In: *Physical Review B* 97.23 (2018), p. 235310.
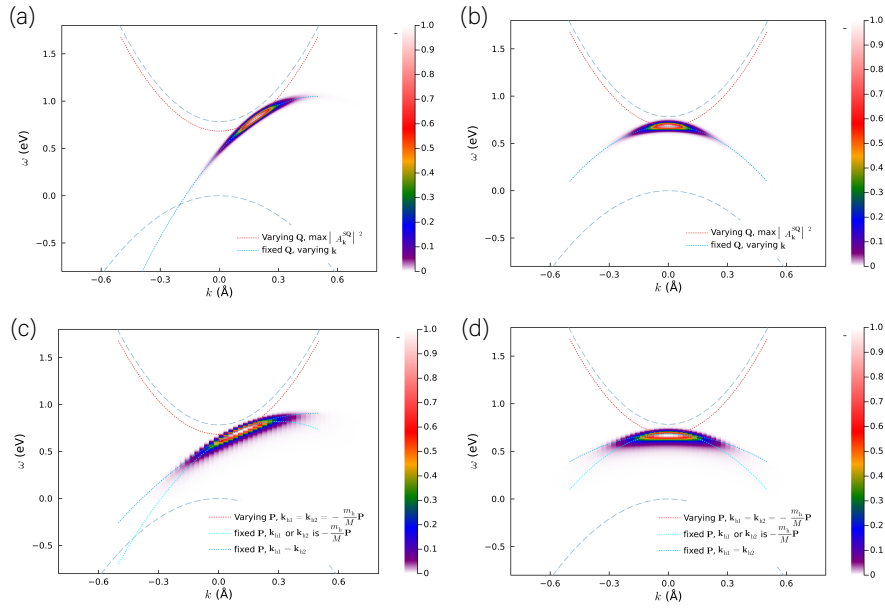
Figure 1: Comparison between exciton signatures and trion signatures. (a), (b) Exciton ARPES signatures at $Q_x = 0\,\text{Å}$ and $0.5\,\text{Å}$. (c), (d) Exciton ARPES signatures at $Q_x = 0\,\text{Å}$ and $0.5\,\text{Å}$. The binding energy of the trion mode is manually shifted to the exciton binding energy in [1], but significant differences can still be spotted between exciton and trion signatures.