Excitonic properties of hBN from a time-dependent Hartree-Fock mean-field theory

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Theoretical description of excitons

We start by considering a many-body system of electron,

$$H_{\text{eq}} = \sum_{\alpha\beta} h_{\alpha\beta} c^{\dagger}_{\alpha} c_{\beta} + \frac{1}{2} \sum_{\alpha\beta\gamma\delta} V^{\alpha\beta}_{\gamma\delta} c^{\dagger}_{\alpha} c^{\dagger}_{\beta} c_{\gamma} c_{\delta}$$

which then we drive out of equilibrium with an externally applied time-dependent force,

$$H_{\text{ext}} = \sum_{\alpha\beta} B_{\alpha\beta} c^{\dagger}_{\alpha} c_{\beta} F(t)$$

We perfom a generic derivation on how the collective excitations emerge from this many-body system of interacting particles within a time-dependent Hartree-Fock mean-field theory. To this end, we study the linear response of the system's reduced density matrix,

$$\rho_{ba}(t) = \left\langle c^{\dagger}_{a}(t) c_{b}(t) \right\rangle$$

in a many-body perturbation theory at zero temperature and demonstrate that it can be expressed in terms of a generalized eigen-problem of the effective two-particle Hamiltonian of the electron-hole interaction,

$$\left(\hbar\omega \begin{bmatrix} \mathbb{1} & 0 \\ 0 & -\mathbb{1} \end{bmatrix} - \begin{bmatrix} \boldsymbol{R} & \boldsymbol{C} \\ \boldsymbol{C}^{\dagger} & \boldsymbol{R}^{*} \end{bmatrix} \right) \begin{bmatrix} \boldsymbol{\rho}^{(1)}_{eo}(\omega) \\ \boldsymbol{\rho}^{(1)}_{oe}(\omega) \end{bmatrix} = \begin{bmatrix} \mathbb{1} & 0 \\ 0 & -\mathbb{1} \end{bmatrix} \begin{bmatrix} \boldsymbol{\mathcal{J}}_{eo}(\omega) \\ \boldsymbol{\mathcal{J}}_{oe}(\omega) \end{bmatrix}$$

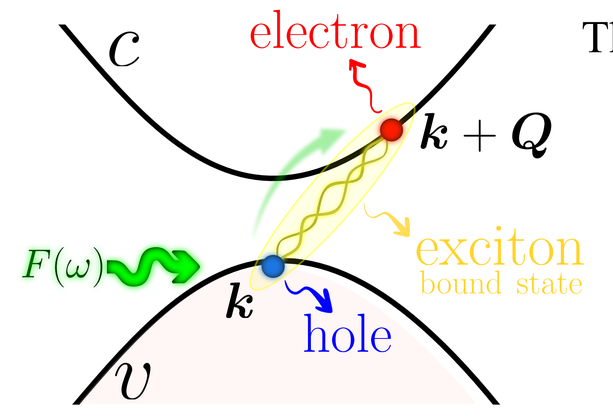
with the resonant and coupling block given respectively by

$$\mathbf{R} \equiv H^{e_3 o_4}_{e_1 o_2} = (\epsilon_{e_1} - \epsilon_{e_2})\, \delta_{e_1 e_3} \delta_{o_4 o_2} + \left(V^{o_4 e_1}_{o_2 e_3} - W^{e_1 o_4}_{o_2 e_3} \right)$$

$$\mathbf{C} \equiv H^{o_3 e_4}_{e_1 o_2} = \qquad 0 \qquad + \left(V^{e_4 e_1}_{o_2 o_3} - W^{e_1 e_4}_{o_2 o_3} \right)$$

Excitonic generalized eigen-problem in a crystal

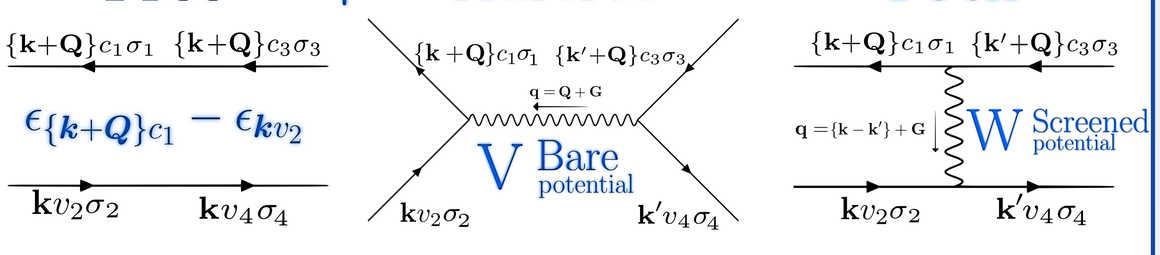
Structure of the generalized eigen-problem in terms of the Bloch momentum and spin degrees of freedom.

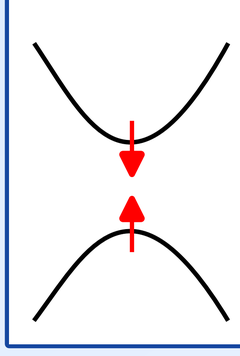


The source term that describes such a perturbation is:

$$\mathcal{J}_{\{k+Q\}c_1, k v_2}(\omega, Q + G)$$

$$H_{\text{e-h}} \overset{\text{TDA}}{\approx} R_{k,k'}(Q)$$





Spin-Singlet

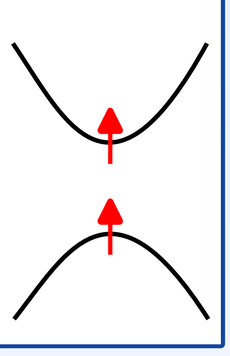
$$R_s = \epsilon - W + 2V$$

controls the details of the excitonic spectrum

Spin-Triplet

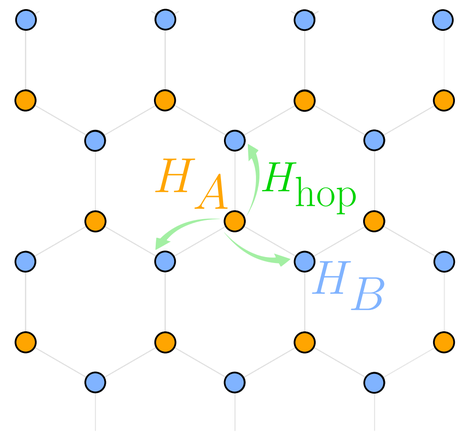
$$R_{\text{t}} = \epsilon - W$$

enables the formation of bound states



hBN eletronic states

The hBN eletronic single-particle energies and wave-functions are obtain within a nearest-neighbour tigh-binding model



$$H_{\text{TB}}(\boldsymbol{k}) = \begin{bmatrix} \epsilon_A & -t\gamma_{\boldsymbol{k}} \\ -t\gamma^{\dagger}_{\boldsymbol{k}} & \epsilon_B \end{bmatrix}$$

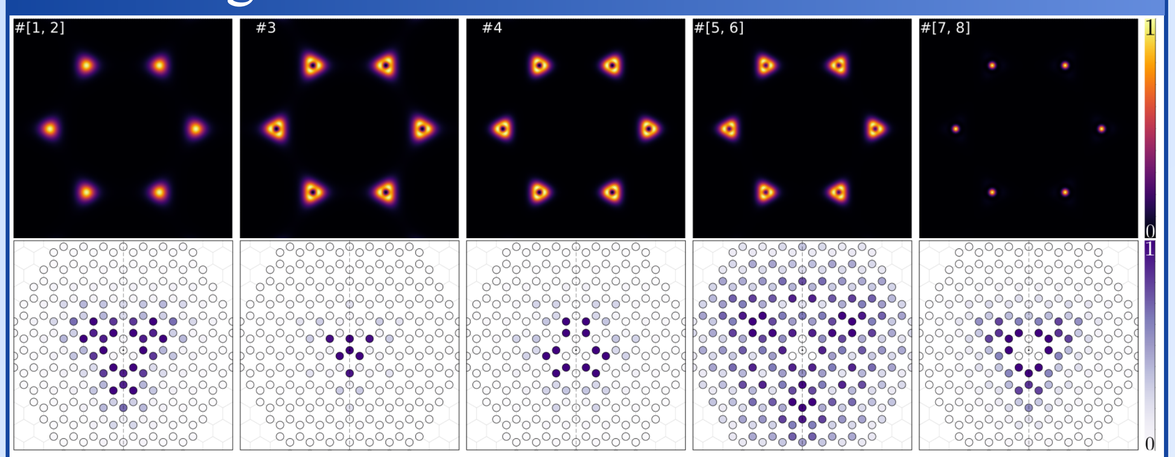
Atomistic e-e interaction

We assume that the electrons are described by ultra-localized Wannier functions such and that they interaction only at long distances

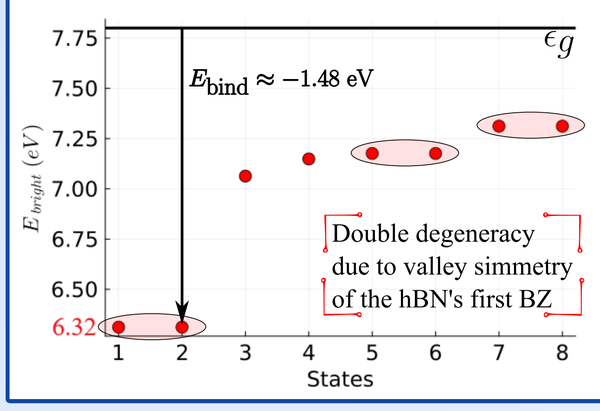
$$V^{\mathbf{k}_1\lambda_1, \mathbf{k}_2\lambda_2}_{\mathbf{k}_3\lambda_3, \mathbf{k}_4\lambda_4} = \frac{1}{\mathcal{V}} \sum_{\mathbf{G}} \int_{1\text{BZ}} \frac{d^2\mathbf{q}}{(2\pi)^2} \mathrm{V}(\mathbf{q}+\mathbf{G})\, \varrho^{\mathbf{k}_1\lambda_1}_{\mathbf{k}_4\lambda_4}(\mathbf{q}+\mathbf{G})\, \varrho^{\mathbf{k}_2\lambda_2}_{\mathbf{k}_3\lambda_3}(-(\mathbf{q}+\mathbf{G}))\, \delta_{\mathbf{q},\{\mathbf{k}_1-\mathbf{k}_4\}} \delta_{\mathbf{q},\{\mathbf{k}_3-\mathbf{k}_2\}}$$

depends explicitly on the single-particle wave functions $\phi^{\zeta}_{\mathbf{k}\lambda}$

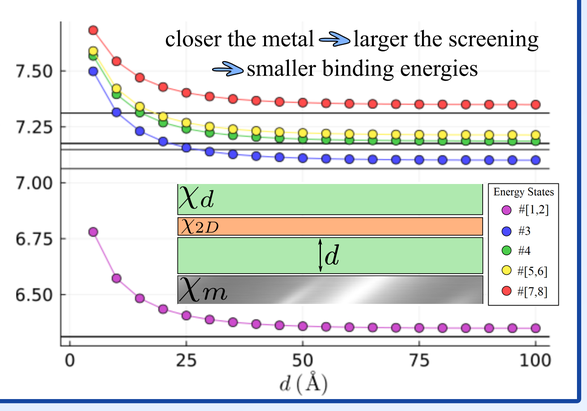
hBN bright exciton states



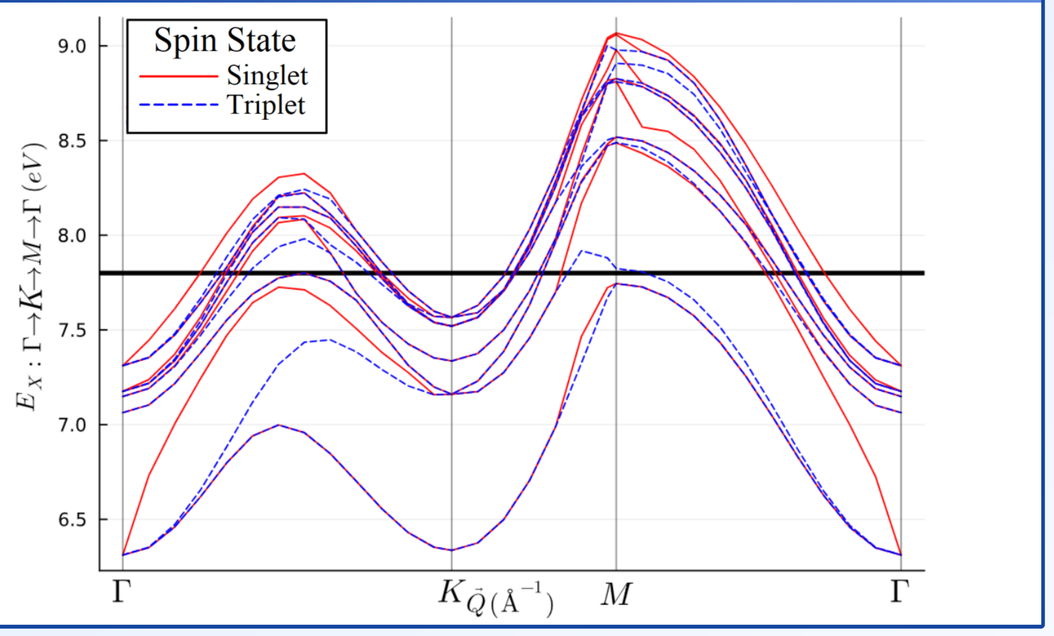
Isolated hBN monolayer



hBN-metal heterostructure



Isolated hBN excitonic band structure



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