Supplemental Material for: Triangular lattice exciton model

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I. SINGLE-PARTICLE MODEL PARAMETERIZATION

The single-particle band structure of the triangular lattice model is

$$\varepsilon_{n\sigma\vec{k}} = \varepsilon_n + \sum_{\vec{\delta}} t_{n\sigma\vec{\delta}}\, e^{-i\vec{k}\cdot\vec{\delta}}, \tag{1}$$

where $n \in \{c, v\}$ and $\sigma \in \{\pm 1/2\}$ are band and spin indices, respectively. Let us center the wave vector $\vec{k}$ at the K point with valley index $\tau \in \{\pm 1\}$ located at $\vec{K}_\tau$ using $\vec{q} \equiv \vec{k} - \vec{K}_\tau$. $\vec{K}_\tau$ along with $\vec{\delta}$ and $t_{n\sigma\vec{\delta}}$ are defined in the main article. The dispersion, its gradient, and Laplacian then become

$$\varepsilon_{n\sigma\tau\vec{q}} = \varepsilon_n + \sum_{\vec{\delta}} t_{n\sigma\vec{\delta}}\, e^{-i\vec{K}_\tau\cdot\vec{\delta}}\, e^{-i\vec{q}\cdot\vec{\delta}}, \tag{2a}$$

$$\nabla_{\vec{q}}\, \varepsilon_{n\sigma\tau\vec{q}} = \sum_{\vec{\delta}} t_{n\sigma\vec{\delta}}\, e^{-i\vec{K}_\tau\cdot\vec{\delta}}\, e^{-i\vec{q}\cdot\vec{\delta}}\, (-i\delta), \tag{2b}$$

$$\nabla^2_{\vec{q}}\, \varepsilon_{n\sigma\tau\vec{q}} = \sum_{\vec{\delta}} t_{n\sigma\vec{\delta}}\, e^{-i\vec{K}_\tau\cdot\vec{\delta}}\, e^{-i\vec{q}\cdot\vec{\delta}}\, (-\delta\cdot\delta), \tag{2c}$$

$$= -\left(\varepsilon_{n\sigma\tau\vec{q}} - \varepsilon_n\right) a^2. \tag{2d}$$

Next, we define $\vec{q} = q\hat{q}$, where $\hat{q} = \hat{x}\cos\alpha + \hat{y}\sin\alpha$. In the limit $q \to 0$, we have

$$\varepsilon_{n\sigma\tau\vec{q}} = \varepsilon_{n\sigma\tau\vec{0}}, \tag{3a}$$

$$\nabla_{\vec{q}}\, \varepsilon_{n\sigma\tau\vec{q}}\Big|_{q\to 0} = 0, \tag{3b}$$

$$\nabla^2_{\vec{q}}\, \varepsilon_{n\sigma\tau\vec{q}}\Big|_{q\to 0} = -\left(\varepsilon_{n\sigma\tau\vec{0}} - \varepsilon_n\right) a^2. \tag{3c}$$

Expanding the dispersion around K_τ in an arbitrary direction α to 2nd order gives

$$\begin{aligned}
\varepsilon_{n\sigma\tau\vec{q}} &\approx \varepsilon_{n\sigma\tau\vec{0}} + \frac{\partial \varepsilon_{n\sigma\tau\vec{q}}}{\partial q_\alpha}\Big|_{q\to 0} q_\alpha + \frac{\partial \varepsilon^2_{n\sigma\tau\vec{q}}}{\partial q^2_\alpha}\Big|_{q\to 0} \frac{q^2_\alpha}{2}, \\
&\approx \varepsilon_{n\sigma\tau\vec{0}} - \left(\varepsilon_{n\sigma\tau\vec{0}} - \varepsilon_n\right) \frac{q^2_\alpha a^2}{4}, \\
&\approx \left(\varepsilon_n - 3t_n + 18\sigma\tau\tilde{t}_n\right) + \left(\frac{3t_n a^2}{4}\right) q^2_\alpha,
\end{aligned} \tag{4}$$

where we have neglected $\tilde{t}_n$ in the q^2_α term and used

$$\frac{\partial \varepsilon_{n\sigma\tau\vec{q}}}{\partial q_\alpha}\Big|_{q\to 0} = \hat{q}\cdot\nabla^2_{\vec{q}}\, \varepsilon_{n\sigma\tau\vec{q}}\Big|_{q\to 0}, \tag{5a}$$

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$$\frac{\partial \varepsilon^2_{n\sigma\tau\vec{q}}}{\partial q^2_\alpha}\Big|_{q\to 0} = \frac{1}{2}\nabla^2_{\vec{q}}\varepsilon_{n\sigma\tau\vec{q}}\Big|_{q\to 0}, \tag{5b}$$

where the last relation follows from isotropy.

Our goal is to have the expanded dispersions above satisfy the effective mass description of the conduction and valence band dispersion given by

$$\varepsilon_c = E_g + \left(\frac{\hbar^2}{2m^*}\right) q^2_\alpha, \tag{6a}$$

$$\varepsilon_v = -\frac{\Delta}{2}\left(1 - 2\sigma\tau\right) - \left(\frac{\hbar^2}{2m^*}\right) q^2_\alpha. \tag{6b}$$

Matching the q^2_α terms with those in Eq. (4) yields $t_c = -t_v = t \equiv 2\hbar^2/3m^*a^2$. Similarly, we get $\tilde{t}_c = 0$ and $\tilde{t}_v = \Delta/18$ from matching the $\sigma\tau$ terms. The remaining terms give $\varepsilon_c = 3t + E_g$ and $\varepsilon_v = -3t - \Delta/2$. To conclude, the six model parameters have been uniquely fixed by the physical quantities E_g, m^*, and Δ.

II. GW CALCULATION DETAILS

The first principles calculations were performed within the GW_0 approximation as implemented in the *Vienna ab initio simulation package*.[1] All calculations used projector augmented wave potentials specifically generated for GW calculations, a $9 \times 9 \times 1$ Γ-centered k-space grid, and a plane wave cut off 320 eV. The geometry relaxation was performed within the local density approximation. The periodically repeated transition-metal-dichalcogenide layers were separated by 8 times the lattice constant. The GW_0 calculations required 4 iterations over the Green functions to reach convergence. The calculations used an energy cut off 200 eV for the response function.

III. MODEL CONVERGENCE

We want to choose a cutoff radius of the grid that is large enough to converge the exciton states of interest. A good estimate is a cutoff radius R_{cut} several times larger that the largest radius of the considered excitons. See Fig. 1. Also note that the largest exciton radii must be converged for this quantity to be meaningful.

IV. MODEL SCALING

As the matrix representation of the model Hamiltonian is sparse, we expect the scaling of the model to be significantly better than the normal $\mathcal{O}(N^3)$ scaling for dense matrices. This expectation is confirmed by Fig. 2 showing that our model scales as $\mathcal{O}(N \log N)$.

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[1] G. Kresse and J Furthmuller, Comput. Mat. Sci. **6**, 15 (1996).

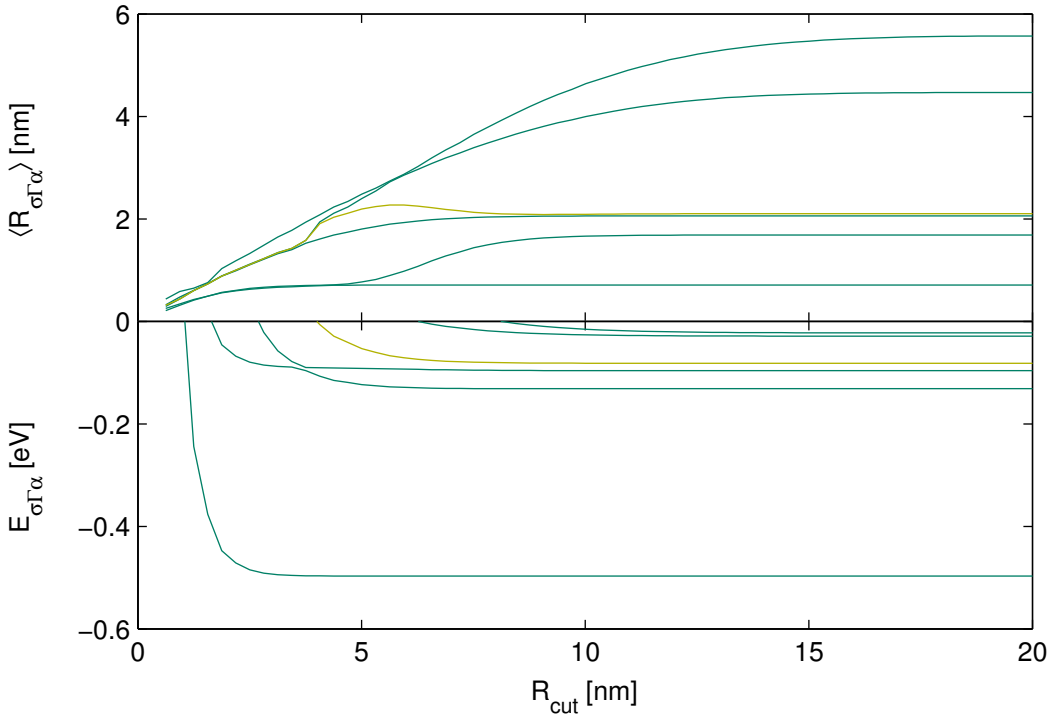

FIG. 1. The average exciton radius and energy as a function of the lattice grid cutoff radius. With 3 of the curves being degenerate, the 6 curves represent the 9 lowest-energy excitons.

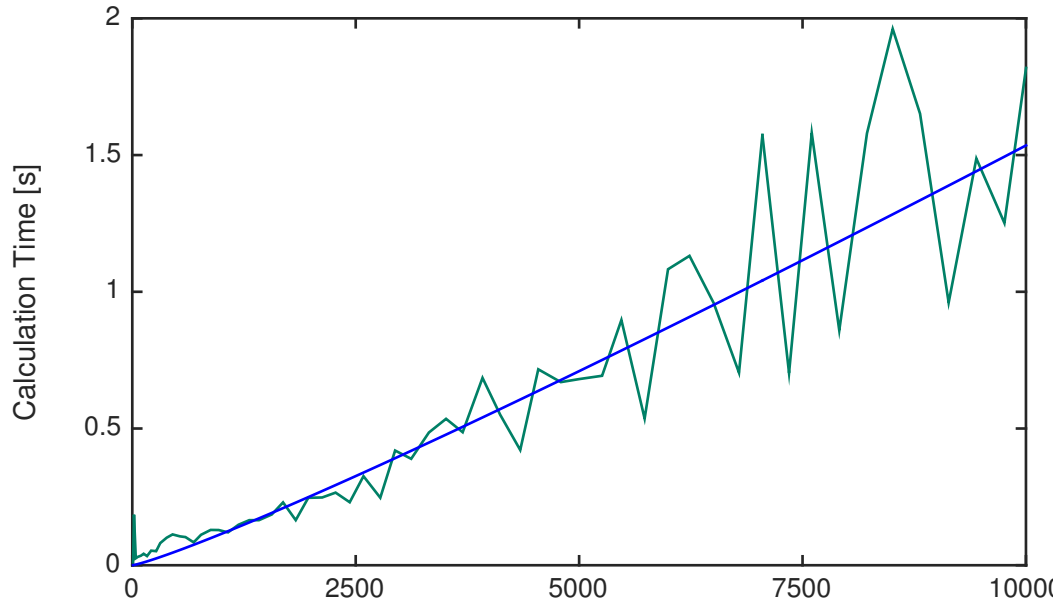

FIG. 2. The computation cost of solving for the 9 lowest exciton levels and states in the 3ALE model is W(N) = k*N*log(N), where N is the number of grid points and $k = (5/3) \times 10^{-5}$ s on a 2.8 GHz Intel Core i7 laptop.