Combined tensor network/cluster expansion method using logic gates:
Illustrated for (bi-)excitons by a single layer MoS$_2$ model system

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Carriers such as electrons and holes inside the Brillouin zone of complex semiconducting materials can form bound states (excitons, biexcitons etc.). For obtaining the corresponding eigenstates (e.g. through Wannier or Bethe Salpeter equation) and dynamics (e.g. cluster expansion) the number of involved electrons and holes as well as the accuracy is limited by the appearing high dimensional tensors (i.e. wavefunctions or correlations). These tensors can be efficiently represented and manipulated via tensor network methods. We show how tensor networks formulated via classic logic gates can be used to treat electron-hole complexes inside the Brillouin zone. The method is illustrated for the exciton and biexciton states of a single layer transition metal dichalcogenide MoS$_2$ like model system.

Semiconductor Bloch-equations and cluster expansion methods have been the work horse for optical induced carrier and exciton dynamics in semiconductor materials for decades [1–7]. Even for the recent monolayer two dimensional materials like monolayer TMDCs, these methods are still successful. However most treatments of Coulomb bound electron-hole states were restricted to exciton states. Trion and biexciton states and beyond are seldom included [4, 8–13]. If they are included, the correlations are expanded in a basis of few bound exciton, trion or biexciton states [4, 8–11, 13]. Exciton states are calculated using the Wannier equation [4, 8, 14–16], either in real space [8, 10, 16, 17] or in reciprocal space [14, 15, 18]. Often the evaluation is restricted for exciton living near high symmetrical points such as the $\Gamma$ or $K$ and $K'$ points. In the context of ab initio treatments the equivalent Bethe-Salpeter equation (BSE) is used for calculating exciton states [18–24]. On the other hand the calculations for higher order correlations like trions or biexcitons are sparse [8, 10, 11, 13, 25–29]. This paper will illustrate a route to make bigger electron-hole complexes accessible: dynamically or for obtaining bound eigenstates.

Higher order correlations (induced by e.g. Coulomb, electron-phonon or electron-photon interaction) for few level systems like quantum dots have been successfully addressed by inductive equation of motion methods [30–32]. However these methods are restricted to systems with few discrete levels and few discrete photon or phonon modes and cannot be applied so far to correlations with many continuous momentum indices. The required memory sizes for storing the correlations scales exponentially in the number of involved particles $N$ and polynomial in the number of involved states $M$ ($\propto M^N$). Even small numbers of involved particles e.g. $N = 4$ lead for a treatment of two dimensional Brillouin zone (BZ) (e.g. for small 30 momentum points in each dimension a two band model yields $M = 1800$) leads to a hard numerical problem. So the stored data required for the simulation is the bottleneck to attack higher order correlations. However the naive raw data amount required to store entire movies on our computers and smartphones is at least impractical, but lossy data compression solves this issue and storing hundreds of movies on a single computer is possible. So for treating higher order many particle correlations a lossy data compression method and the ability to calculate directly on the compressed data will be the solution. Expanding the correlations in a basis (e.g. exciton [4, 9], trion [10, 11, 25], biexciton [13, 26], permutational symmetric basis [33, 34]) is in principle already a first simple form of data compression, where known symmetries and properties of the problem are used for an efficient description of the system. However for every problem a different or modified basis is required, where the reformulation and implementation of the equations is tedious and requires substantial effort. In the context of highly correlated quantum systems tensor network methods like matrix product states (MPS) provided a systematic and reliable way to store and manipulate quantum states of e.g. spin chains [35–38], and also system bath interactions [39, 40]. The wavefunction of the spin chain is interpreted as a tensor and decomposed in a tensor network such as a MPS (also called a tensor train (TT)). In mathematics and chemistry a new trend uses TT (or other tensor networks) to compress high dimensional tensors regardless, if the tensor represents an actual quantum mechanical wavefunction [40]. Furthermore for solving partial differential equations in real space quantics tensor trains (QTT) were introduced [41–46]. QTT do not use the spatial coordinates as indices of the tensors, but their binary representation. We transfer this concept to cluster expansion and Wannier equations, since correlations appearing there are also tensors. We show that a binary representation of the BZ quasi momentum allows a straightforward expression of the material equations using tensor networks with binary logic gates. We focus on the calculation for a
model system with realistic numerical complexity, that describes excitons and biexcitons formed between valence and conduction band of the two dimensional TMDC MoS$_2$ [8, 13, 15, 27, 47–52]. We demonstrate that the electrons and holes complex quantities can be calculated using a very high number of grid points in the BZ. This paper is a proof of principle, that the combination of tensor network methods increases the range of problems addressable with Wannier equation and BSE. Beside the calculation of (bi-)exciton states the concept is also extendible towards solving equations of motions in cluster expansion.

**Model System:** The Hamilton operator $H$ of the model system is $H = H_0 + H_C$. The electronic bandstructure enters the Hamiltonian through $H_0 = \hbar \sum_{k,\lambda} \varepsilon_k^\lambda a_{k\lambda}^\dagger a_{k\lambda}$, here $k$ is the quasi momentum in BZ, and $\lambda$ describes band and spin. $\varepsilon_k^\lambda$ is the bandstructure of the material, for this paper the tight binding bandstructure for MoS$_2$ from [53, 54] is used. $a_{k\lambda}^\dagger$, $a_{k\lambda}$ are the creation and annihilation operator of an electron or hole, distinguished by $\lambda$. The Coulomb interaction Hamiltonian $H_C$ reads

$$H_C = \sum_{k_1, k_2, q_1, q_2, \lambda_1, \lambda_2} \frac{1}{\lambda_1 \lambda_2} \varepsilon_{k_1}^{\lambda_1} \varepsilon_{k_2}^{\lambda_2} \langle a_{k_1 q_1}^\dagger a_{k_2 q_2} \rangle,$$

$C_{\lambda_1}^{\lambda_2}$ is 1, if $\lambda_1$ and $\lambda_2$ are both holes or both electron and -1 otherwise. The prefactor $I_{\lambda_1, \lambda_2}^{k_1, k_2} = F_{q_1 q_2} V_{q_1 q_2}^{\lambda_1 \lambda_2}$ includes the Keldysh style Coulomb potential $V_q$ [15, 55] and the tight-binding (TB) coefficients $\kappa_{n_1}$ inside

$$F_{q_1 q_2}^{k_1 k_2} = \sum_{n_1, n_2} C_{k_1 n_1, k_2 n_2} C_{k_2 q_1 + q_2} C_{k_1 - C_{\lambda_1}^{\lambda_2} q_{n_1}}.$$

The model system is slightly simplified (no exchange coupling term), since this paper is focused on the method (more accurate treatments are subject to future studies). We introduce the multiindex $k = (k \lambda)$, $\lambda$ is only written explicitly, if needed. The correlations describing the system are $(a_{k^1}^\dagger ... a_{k^n}^\dagger a_{k^1} ... a_{k^n}^\dagger) = S(k^1 ... k^n | k^1_R ... k^n_R)$. Using Heisenberg equations of motion $\partial_t \langle O \rangle = i/\hbar \{[H, O], -\rangle$ we arrive an equation for $S$:

$$\partial_t S(k^1 ... k^n | k^1_R ... k^n_R) = i \sum_{j=1}^n \varepsilon_j k_j S(...)$$

$$-2i \sum_{j, q} I_{\lambda^1 \lambda^2}^{k^1 q} S(...) k^1 + q | k^1_R - C_{\lambda^1}^{\lambda^2} q | k^1_R k$$

$$-2i \sum_{i < j, q} I_{\lambda^1 \lambda^2}^{k^i q} S(...) [k^i_R - C_{\lambda^1}^{\lambda^2} q] | k^i_R + q | k$$

$$\{L \leftrightarrow R, n \leftrightarrow m\}. \tag{1}$$

On the rhs only the changes in the indices of $S(...) | k$ compared to the lhs are denoted. Note, some terms on rhs change the number of indices of $S(...) | k$ compared to the lhs. Eq. (1) creates the usual infinite hierarchy of correlations. Examples of higher order correlations include important spectroscopic contributions such as $S(k_1 k_2 k_3 | k_1)$ (contains density assisted polarizations leading to excitation induced dephasing), $S(k_1 k_2 k_3 k_i)$ (contains biexcitonic coherences) or $S(k_1 k_2 k_3 k_i | k_5 k_6)$ (contains single exciton to biexciton correlations). These higher order tensors $S(\ldots | \ldots)$ impose the high numerical burden, that tensor network methods will lift. While for the future, calculation of the quantum dynamics using Eq. (1) using the tensor network approach are possible, we focus in this paper on the calculation of many particle eigenstates, i.e. eigenstates for excitons, biexcitons etc.. Like in the calculation of the Wannier equation [14], we take the homogenous part of Eq. (1) and convert the equation to an eigenproblem with eigenenergies $E$ for the respective many particle complexes:

$$iE S(k^1_L ... k^n_L | k^1_R ... k^n_R) = \sum_{j=1}^n \varepsilon_j S(...)$$

$$-2i \sum_{i < j} I_{\lambda^1 \lambda^2}^{k^i q} S(...) [k^i_R - C_{\lambda^1}^{\lambda^2} q] | k^i_R + q] S(...) k$$

$$\{L \leftrightarrow R, n \leftrightarrow m\}. \tag{2}$$

For $S(k_1 k_2)$ Eq. (2) corresponds to the Wannier equation for excitons (bound electron hole pairs) [14] in reciprocal space and is equivalent to a BSE [4, 21, 28]. For $S(k_1 k_2 k_3 k_4)$ Eq. (2) is the generalization to biexcitons, i.e. bound complexes from two electrons and two holes. In this paper, we will focus on excitons and biexcitons.

**Tensor Network Methods:** The tensor $S(k^1_L ... k^n_L | k^1_R ... k^n_R)$ is a tensor with indices $k^1_L, \ldots, k^n_L, k^1_R, \ldots, k^n_R$ and rank $n + m$. If we assume $g = 1000$ grid points for the BZ (which is probably too small), the memory requirement is $g^{n+m} = 1000^{n+m}$, so that already for very small $n$ and $m$ the memory exceeds the feasible and possible range. [56] showed that every tensor $T_{k_1 \ldots k_N}$ can be approximated as MPS (in mathematics TTT) in the form:

$$T_{k_1 \ldots k_n} = \sum_{\alpha_1 \ldots \alpha_n} \Gamma^T_{\alpha_1 \alpha_2} \Gamma^T_{\alpha_2 \alpha_3} \ldots \Gamma^T_{\alpha_{n-1} \alpha_n}.$$

The tensors $\Gamma_{\alpha_{\alpha'}}$ have a maximum of $g \cdot d^2$ elements, if $d$ is the maximum number of $\alpha$ (link dimension). If the relevant information of the tensor can be represented with small $d$, the overall memory size reduces from exponential scaling $g^{n+m}$ to linear scaling $(n + m) \cdot g \cdot d^2$ making higher dimensional tensors accessible [37, 56–58]. In the following we will use a diagrammatic notation for tensors [37, 57, 58]: The tensor is represented by a
of index information from the initial MPS (S the key design principle is to ensure the correct flow of information from the initial MPS (S) to the term prefactors inside the MPO to the final MPS indices (S(...|...)) on the rhs). In the TN in Fig. 2a) the connections and junction ensure that the same indices of the initial tensor on the rhs of Eq. (2), the energy tensor εkR and the tensor on the lhs are connected.

Constructing the TN for the Coulomb term −2i∑q,α,β∈J S(0) · kR a S(...|...){kR − Cλ q}...{kR + q} is more involved and will require five MPOs, which are subsequently compressed to a single MPO. q has positive and negative components, negative number are encoded using two’s complement representation for binary negative integers [60], which matches nicely the periodic properties of the BZ. (A negative q is represented by a positive q + G inside BZ with suitable G.) For the correct flow of information the TN has to connect the bit indices for kR, kR, kR − q, kR + Cλ q and q with Cλ = ±1. 

Tensors representing binary logic gates achieve this: a set of full adders [60] calculate kR − q and kR + Cλ q from kR, kR and q. For the case Cλ = −1 additional NOT circuits convert q to a negative input in two’s complement representation for the full adder. The corresponding TN is shown in Fig. 2c), the application of Vq and calculation of the indices is handled by the MPO in the middle of the set of five MPOs. Here the fulladders combine the k indices and the q indices of Vq for every bit of the binary representation. In addition carry bits connect

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\begin{align*}
\text{Figure 2. a) TN representing the homogeneous part of Eq. (2), b) full adder (FA) and negation (NOT) logical circuits, c) TN representing the Coulomb term of Eq. (2) (only k part).}
\end{align*}
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the full adder for different bit digits. In Fig. 2c) the prefactor $\mathcal{F}_{\mathbf{k}}^{k_R,k_L}$ is handled by the four outer MPO’s. For including it, the MPO from Fig. 2 is combined with two MPO’s representing $c_{\mathbf{k}+\mathbf{q}+\mathbf{q}_2}\mathcal{F}_{\mathbf{k}_1}^{\mathbf{k}_1}-c_{\mathbf{k}_1}^{\mathbf{k}_1}\mathcal{F}_{\mathbf{k}_2}^{\mathbf{k}_2}$ before its application and two MPOs representing $c_{\mathbf{k}_R}^{\mathbf{k}_R+n_1}c_{\mathbf{k}_L}^{\mathbf{k}_L+n_2}$ after its application. (Supplemental material includes a more extensive discussion).

We use the ITensor C++ library (patched version 2.1.0), for the calculation of all tensor operations [61]. The rhs of Eq. (2) is calculated through the TN brought in the form of an MPO, compressed using a fitApply algorithm. In order to solve Eq. (2) and to determine the respective exciton and biexciton eigenenergies and wavefunctions, a density matrix renormalization group (DMRG) algorithm [58, 62] is used which is capable of obtaining the eigenvalues and eigenvectors (MPS) of a MPO. We use a modified DMRG algorithm based on the itensor DMRG [61] implementation for multiple MPO and for calculation of higher energy eigenvectors. Thus, Eq. (2) is solved for the exciton $S(k^+_1 k^+_2)$ and biexciton $S(k^+_R k^+_L k^+_1 k^+_2)$ coherences/wavefunction on a full 1024 × 1024 grid for every k-vector inside the full BZ. To address optical excitability states, we focus on (bi-)exciton states with zero overall momentum $\langle a^+_1 \alpha_1 a^+_2 \lambda_2 \rangle$ and $\langle c^+_1 \lambda_2 a^+_1+\mathbf{q}_2 a^+_1-\mathbf{q}_2 \lambda_2 a^+_1 \lambda_2 \rangle$, the TN constructing this coherence from $S(k^+_R k^+_L k^+_1 k^+_2)$ is given in the Supplemental material.

We obtain the bound, bright (dark) A and B exciton at 1.798 eV (1.794 eV) and 1.939 eV (1.943 eV) compared to a band gap of 2.124 eV at the K-point, reproducing [54], whose band structure [53, 54] is used in the model system, here. As an example from the exciton states, Fig. 3(a) shows the A exciton wavefunction (1.798 eV) for parallel spin up configuration localized at the K-valley. The biexciton coherence with zero center of mass momentum $\langle c^+_1 \lambda_2 a^+_1+\mathbf{q}_2 a^+_1-\mathbf{q}_2 \lambda_2 a^+_1 \lambda_2 \rangle$ depends on the three momenta $\mathbf{k}$, $\mathbf{k}'$ and $\mathbf{q}$. To characterize the six dimensional wavefunction, we sum over two momenta (e.g. $\mathbf{k}'$ and $\mathbf{q}$) while plotting over the third (e.g. over $\mathbf{k}$) in the BZ. Fig. 3(c)-(h) shows two example biexciton states: Two A excitons, one electron-hole pair with parallel spin up located at the K- (Fig. 3(c)) and another with parallel spin down at the K’-valley (Fig. 3(e)), constitute a bright biexciton (note $\mathbf{q} \approx 0$, Fig. 3(g)) with an energy of 3.563 eV (33 meV binding energy, cf. [47, 51]).

Furthermore, the approach allows also to access many higher energy bound biexciton states (bright or dark). Fig. 3(d),(f) and (h) shows as an example a biexciton composed from two dark 3d-excitons with anti-parallel electron-hole spin, (see wavefunction depicted in Fig. 3(b), cf. [63]). Beside the example exciton and biexciton states, the framework allows to determine many higher energy bound electron holes states, in principle also for other correlated electron and hole quasi-particles like tritons.

In conclusion, the combination of tensor networks, cluster expansion and logic gates on the Brillouin zone allows to easily access bound electron hole quasi-particle with little numerical cost and high precision. Future studies in this framework will provide systematic investigation of the bound electron-hole complexes and extend the numerical technique to quantum dynamics.

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Figure 3. Selected exciton and biexciton wavefunctions in BZ: (a) bright A exciton with parallel spin up and (b) dark 3d exciton with anti parallel spin localized at the K-valley. (c), (e) and (g) bright biexciton formed from two bright A excitons and (d), (f) and (h) dark biexciton formed from two dark 3d excitons. The plotted variable is depicted in the corner. (g) and (h) are plotted using a logarithmic color scale.
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