


Modeling excitonic Mott transitions in two-dimensional semiconductorsA. Kudlis and I. Iorsh *Department of Physics and Technology, ITMO University, St. Petersburg 197101, Russia*

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We analyze the many-particle correlations that affect the optical properties of two-dimensional semiconductors. These correlations manifest themselves through the specific optical resonances such as excitons, trions, etc. Starting from the generic electron-hole Hamiltonian and employing the microscopic Heisenberg equation of motion the infinite hierarchy of differential equations can be obtained. In order to decouple the system we address the cluster expansion technique which provides a regular procedure of consistent accounting of many-particle correlation contributions into the interband polarization dynamics. In particular, the partially taken into account three-particle correlations modify the behavior of absorption spectra with the emergence of a trion-like peak additional to excitonic ones. In contrast to many other approaches, the proposed one allows us to model the optical response of 2d semiconductors in the regime when the Fermi energies are of the order of the exciton and trion binding energies, thus allowing us to rigorously model the onset of the excitonic Mott transition, the regime being recently studied in various 2d semiconductors, such as transition metal dichalcogenides.

DOI: [10.1103/PhysRevB.103.115307](https://doi.org/10.1103/PhysRevB.103.115307)**I. INTRODUCTION**

Two-dimensional semiconductors, such as monolayers of transition metal dichalcogenides (TMDs), appear to be an ideal platform for the exploration of the excitonic complexes [1]. Peculiar and appealing properties of TMDs are largely dictated by their two-dimensional nature: suppressed screening leads to the emergence of the tightly bound [2–4] and at the same time strongly interacting excitons in these structures [5,6]. The former property allows for the efficient optical probing of the exciton structure [1,7–10], and latter leads to the emergence of the pronounced many-particle correlations. Besides that, the 2d nature of TMDs allows for the efficient doping of these structures by means of an external gating [11]. The most pronounced effect arising in doped TMDs is the formation of the additional peak in photoluminescence redshifted with respect to the excitonic one [12–16].

From the theoretical side, there still exists an ambiguity in the interpretation of the origin of this peak. While some of the researchers describe the peak as trions, tightly bound complexes of two electrons and a hole (or vice versa) [17–24], the other part of the community exploits the approach based on the Fermi polarons: excitons, dressed by the sea of the residual electrons [25–29]. Although it has been shown that the two approaches are equivalent in the low doping limit [30], they produce qualitatively different results in the large doping limit [31]. Moreover, at elevated doping and thus electron concentration the composite nature of the exciton should be taken into account: ultimately, as the average separation between the electrons is of the order of the exciton Bohr radius, the excitons can be no longer treated as bound pairs which results in the onset of the exciton Mott transition, where the exciton gas transforms to the electron-hole plasma. The Mott transition in TMD monolayers (MLs) has been recently

explored experimentally [32]. The quantitative model of high-density behavior of the trions in TMDs is still yet to appear. Moreover, the experimentally relevant regime of the excitonic Mott transition [33] still lacks adequate quantitative theoretical description.

One of the methods proved powerful for the description of the excitons in the vicinity of the Mott transition is based on the so-called cluster expansion technique [34–36]. Within this approach, we start with the many-particle Hamiltonian of electrons interacting with the classical time-dependent electromagnetic field. This quantum problem allows for the reformulation in terms of an infinite series of coupled differential equations for many-particle correlations. The cluster expansion defines the scheme of the truncation of this system to obtain the many-body dynamics with controlled accuracy. The cluster expansion technique has been used to quantitatively model the excitonic Mott transition in GaAs quantum wells [37,38].

It has however never been used to model the high-density trion dynamics due to both the computational complexity of the resulting equations and the weakness of the trion response in GaAs quantum wells.

In this paper, we apply the cluster expansion technique for the TMD MLs to explore the effect of the high-order many-particle correlations on the optical response of these structures (see Fig. 1). By extending the system of equations to capture three-particle contributions we were able to extract the emergence of the trion peak in the optical polarizability of TMDs starting barely from the electron-hole Hamiltonian. We believe that the results of the paper prove that the cluster expansion methods could be extremely useful to study the dynamical many-body correlations in TMDs and their effect on the transient optical properties.

The paper is organized as follows. In Sec. II we describe the model and a set of approximations adopted within this work. In Sec. III we show the approach used and also the equation of motion (EOM) for different orders. The final analytical expressions and the corresponding numerical results are presented and discussed in Sec. IV. Finally, in Sec. V we will draw a conclusion.

Throughout the work we try to use indices i, j as summation ones, while primed indices— i', j' —will be used as external ones. Also, if some quantity is time-dependent, we can demonstrate it as $Q(t)$, Q_t , or Q^t . In this case, the corresponding Fourier transform is denoted by $Q(\omega)$, Q_ω , or Q^ω , respectively.

II. DESCRIPTION OF THE MODEL

In what follows we limit ourselves to the case of just two bands. The generalization to the case of the multiband structure is straightforward. We also neglect the spin degree of freedom and consider the single-valley dynamics, thus neglecting the intervalley scattering. Finally, we do not consider the electron-phonon interaction in this work. Thus, the model Hamiltonian reads

$$\hat{H} = \hat{H}_0 + \hat{V}, \quad \hat{V} \equiv \hat{H}_{el,p}, \quad \hat{H}_0 = \hat{H}_{el,k} + \hat{H}_I, \quad (1)$$

where

$$\hat{H}_{el,k} = \sum_{\lambda,k} E_{\lambda,k} \hat{a}_{\lambda,k}^\dagger \hat{a}_{\lambda,k} = \hbar \sum_i \varepsilon_i \hat{a}_i^\dagger \hat{a}_i, \quad \hat{H}_I \approx - \sum_k \mathcal{E}(t) d_{cv} (\hat{a}_{c,k}^\dagger \hat{a}_{v,k} + \hat{a}_{v,k}^\dagger \hat{a}_{c,k}) = \hbar \sum_{i,j} h_{ij}^{\text{ext}}(t) \hat{a}_i^\dagger \hat{a}_j, \quad (2)$$

$$\begin{aligned} \hat{V} &= \frac{1}{2} \sum_{\substack{k_1, k_2 \\ q \neq 0}} V_q [\hat{a}_{c,k_1+q}^\dagger \hat{a}_{c,k_2-q}^\dagger \hat{a}_{c,k_2} \hat{a}_{c,k_1} + \hat{a}_{v,k_1+q}^\dagger \hat{a}_{v,k_2-q}^\dagger \hat{a}_{v,k_2} \hat{a}_{v,k_1} + 2\hat{a}_{c,k_1+q}^\dagger \hat{a}_{v,k_2-q}^\dagger \hat{a}_{v,k_2} \hat{a}_{c,k_1}] \\ &= \frac{1}{4} \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2} \hat{a}_{j_1}, \quad \text{where } i = (\lambda, \mathbf{k}), \quad \lambda \in \{c, v\}. \end{aligned} \quad (3)$$

Indices c and v correspond to conduction and valence bands, respectively. For single-particle kinetic energies we turn to the parabolic isotropic dispersion relation. $\mathcal{E}(t)$ is the electric field strength, while d_{cv} is effective dipole matrix element of interband absorption. As usual, the creation and annihilation

operators obey the following commutation relations:

$$\begin{aligned} [\hat{a}_{j_1}^\dagger, \hat{a}_{j_2}^\dagger]_+ &= 0, \quad [\hat{a}_{i_1}, \hat{a}_{i_2}]_+ = 0, \\ [\hat{a}_{j_1}^\dagger, \hat{a}_{i_1}]_+ &= \delta_{i_1, j_1} \equiv \delta_{\lambda_1, \mathbf{k}_1; \lambda_2, \mathbf{k}_2} \equiv \delta_{\mathbf{k}_1, \mathbf{k}_2} \delta_{\lambda_1, \lambda_2}. \end{aligned} \quad (4)$$

For a greater versatility for each term of the Hamiltonian one can introduce the tensor functions h_{ij}^{ext} and v_{i_1, i_2, j_1, j_2} . The connection formulas between them and standard notation can be found in Appendix B. Note only here that the *small potential*— v_{i_1, i_2, j_1, j_2} —has to satisfy the following symmetry conditions:

$$v_{i_1, i_2, j_1, j_2} = -v_{i_2, i_1, j_1, j_2}, \quad v_{i_1, i_2, j_1, j_2} = -v_{i_1, i_2, j_2, j_1}. \quad (5)$$

It should be marked also that during the derivation of all basic expressions we do not specify the form of these functions. Hence, any model reducible to this structure can be treated by the approach used in this work. Moreover, this formalism can be applied to systems with bosons.

Speaking about the TMD MLs, when it comes to specific numerical calculations, the conventional Coulomb potential in two dimensions ($\sim 1/q$) is no longer applicable [39–41]. There are two reasons of its deviation and both of them relate to screening phenomena. The first modification is associated with the dynamical screening caused by the presence of free carriers. The frequency and momentum dependencies of this effect often lead to difficulties of theoretical description. In a such situation it is necessary to exploit some approximation. In particular, within the random-phase approximation (RPA) the standard Coulomb potential is replaced with some effective one given by the Lindhard formula [38,42].

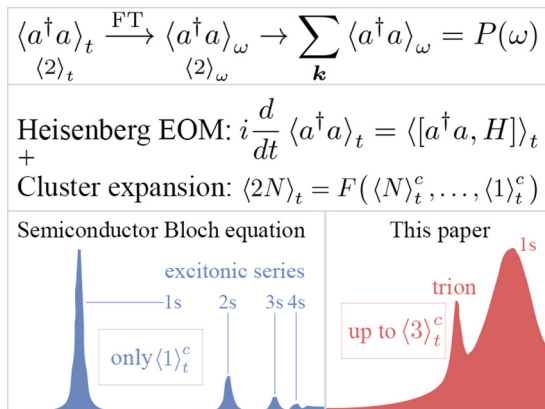


FIG. 1. Schematic representation of the procedure employed in the paper. The key quantity within the work is the interband polarization $P(\omega)$ which allows us to calculate the optical absorption. The former in turn can be expressed via two-operator expectation values $\langle 2 \rangle_t$ which enter the hierarchy of differential equations. The Heisenberg equation together with the cluster expansion technique allows us to regularly truncate such infinite system within the chosen order of correlations $\langle N \rangle_t^c = \langle \hat{a}_1 \dots \hat{a}_N \hat{a}_N^\dagger \dots \hat{a}_1^\dagger \rangle_t$. In this paper we partly take into account the dynamics of three-particle correlations $\langle 3 \rangle_t^c$ thereby expanding the solution given by the semiconductor Bloch equation.

Schematically, it can be expressed as

$$V_q \rightarrow V_q^{\text{eff}} = \frac{V_q}{1 - V_q \Pi(\mathbf{q}, \omega)}, \quad (6)$$

where $\Pi(\mathbf{q}, \omega)$ is free-particle polarization, described by

$$\Pi(\mathbf{q}, \omega) = \sum_k \frac{f_{k-q} - f_k}{\hbar(\omega + i\delta + \varepsilon_{k-q} - \varepsilon_k)}, \quad (7)$$

where f_k is the Fermi-Dirac distribution function. Within the present paper we expect that effects connected with screening caused by free carriers are incorporated automatically by taking into account many-particle correlations, and its artificial injection into the theory via different effective potentials is excessive. The only thing which we assume is that the dynamical screening is fully developed and we deal with stationary systems.

The second type of screening effect is associated with dielectric properties of both substrate, superstrate, and environment which are always present in realistic physical experiments. Due to the dimensional confinement in MLs any inhomogeneity of surroundings leads to a significant distortion of the 2d Coulomb law in the layer. By solving the Poisson equation in such compound structure one can obtain some potential for MLs of a finite thickness [43]. Within the strict two-dimensional limit, however, a such potential tends to the well-known Rytova-Keldysh form [44–47]. The Fourier transform of it is as follows:

$$V_q = \frac{2\pi e^2}{L^2} \frac{1}{\varepsilon(q)q}, \quad (8)$$

where $\varepsilon(q) = \varepsilon_0(1 + qr_0)$. The characteristics ε_0 and r_0 can be considered as phenomenological parameters of the theory. They correspond to the average dielectric constant of surroundings and effective screening length. As was just mentioned, the expression (8) was obtained taking the strict 2d confinement into account. Such approximation is applicable if the typical radius of the bonded electron-hole pairs exceeds the lattice constant, which is realized in the case of TMD MLs. Moreover, the Rytova-Keldysh potential turns out to be well applicable in order to analyze the formation of dipolar excitons and accompanied phenomena in double-layer heterostructures with using the different TMD MLs [48]. Thus, as a final potential used in our numerical calculations we take the expression (8).

As mentioned above, we aim at calculating the interband polarization, which can be written as

$$P(t) = \sum_{\lambda, \lambda', k} \langle \hat{a}_{\lambda, k}^\dagger \hat{a}_{\lambda', k} \rangle_t \mathbf{d}_{\lambda, \lambda'} = \sum_{\lambda, \lambda', k} P_{\lambda \lambda', k}(t) \mathbf{d}_{\lambda \lambda'}. \quad (9)$$

Taking into account the two-band approximation adopted in this work, it is necessary to consider only the following quantity:

$$\tilde{\mathfrak{P}}_k^t \equiv \tilde{\mathfrak{P}}(\mathbf{k}, t) \equiv P_{vc, \mathbf{k}}(t) = \langle \hat{a}_{v, \mathbf{k}}^\dagger \hat{a}_{c, \mathbf{k}} \rangle_t. \quad (10)$$

However, further only due to the computational reasons we prefer to work with the following variable:

$$\mathfrak{P}_k^t \equiv \mathfrak{P}(\mathbf{k}, t) = \langle \hat{a}_{c, \mathbf{k}} \hat{a}_{v, \mathbf{k}}^\dagger \rangle_t \equiv \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t = -\tilde{\mathfrak{P}}_k^t, \quad (11)$$

where we used compound indices: $j' = (v, \mathbf{k})$, $i' = (c, \mathbf{k})$. The transition from one variable to another is trivial.

III. CLUSTER EXPANSION

As was previously mentioned, during the derivation of dynamical equations for quantities of interest we are faced with the infinite system of equations. In this section, we describe the procedure of the proper truncation which makes the system closed. Also, we present the series of approximations for the part which we treat numerically.

A. Equation of motion

In order to describe the time evolution of the arbitrary operator \hat{A} one can address the microscopic Heisenberg equation of motion (EOM). In terms of the model (1) it reads

$$\hbar \frac{d}{dt} \hat{A} + i[\hat{A}, \hat{H}_{el, k}] + i[\hat{A}, \hat{H}_I] = -i[\hat{A}, \hat{V}]. \quad (12)$$

The same is true for the expectation value of operator \hat{A} with an initial statistical operator ρ_0 at an initial time: $\langle A \rangle_t = \text{Tr}[\rho_0 A(t)]$. Speaking about ρ_0 there are no special restrictions on its structure.

Further, we focus on the particular type of the operator \hat{A} for which all the expressions presented in this paper are valid. The corresponding general form can be written as

$$\hat{A} = \hat{a}_{i'_1} \dots \hat{a}_{i'_n} \hat{a}_{j'_n}^\dagger \dots \hat{a}_{j'_1}^\dagger. \quad (13)$$

It is clear that with such operator in hand in the noninteracting case ($V = 0$) from (12) we obtain a closed system of dynamical equations which contains only one type of expectation value. A completely different situation, however, is observed if we are dealing with nonzero potential. In particular, the dynamics of the two-operator expectation value— $\langle \hat{a}_i^\dagger \hat{a}_{i'} \rangle_t$ —is coupled with four-operator ones. The latter, in their turn, depend already on six-operator expectation value dynamics and so on. These steps lead us to infinite hierarchy of differential equations. In order to solve this system, one should find a proper way of truncation of this system. One of the options is factorization of many-operator expectation values into the product of dominant two-operator terms. This procedure results in the RPA that, unfortunately, does not allow one to tackle multiparticle effects. To overcome this problem in [36] some approach on the basis of the cluster expansion technique (CET) presented in [35] was suggested. The main idea is to rewrite all differential equations on expectation values in terms of correlations. By means of the CET the expectation value of the product of an arbitrary combination of creation and annihilation operators \hat{b}_i can be expanded as follows:

$$\begin{aligned} \langle b_1 \rangle_t &= \langle b_1 \rangle_t^c, \\ \langle b_1 b_2 \rangle_t &= \langle b_1 b_2 \rangle_t^c + \langle b_1 \rangle_t^c \langle b_2 \rangle_t^c, \\ \langle b_1 b_2 b_3 \rangle_t &= \langle b_1 b_2 b_3 \rangle_t^c + \langle b_1 b_2 \rangle_t^c \langle b_3 \rangle_t^c + \langle b_2 \rangle_t^c \langle b_1 b_3 \rangle_t^c \\ &\quad + \langle b_1 \rangle_t^c \langle b_2 b_3 \rangle_t^c + \langle b_1 \rangle_t^c \langle b_2 \rangle_t^c \langle b_3 \rangle_t^c, \dots \end{aligned} \quad (14)$$

For the n th-order correlations the sum extends over all disjoint partitions of the set $\{b_1, \dots, b_n\}$. These expressions can be considered as the definition of correlations. This means that in order to obtain the n -operator correlation we have to subtract

from the n -operator expectation value all the lower-order correlations ($n - 1, n - 2, \dots$). The operators in each correlation retain their order. The sign of each term is defined by the number of permutations of fermionic operators in order to coincide with the initial one.

Within this work we require the conservation of the fermionic occupation number. This automatically causes that correlations for odd numbers of operators vanish as well as for combinations where numbers of annihilation and creation operators do not coincide. In particular, for two-operator expectation values we have

$$\langle \hat{a}_i \hat{a}_{j'}^\dagger \rangle_t = \langle \hat{a}_i \hat{a}_{j'}^\dagger \rangle_t^c, \quad \langle \hat{a}_i^\dagger \hat{a}_{j'} \rangle_t = \langle \hat{a}_i^\dagger \hat{a}_{j'} \rangle_t^c, \quad (15)$$

while for four-operator ones the corresponding expansion reads

$$\begin{aligned} \langle \hat{a}_i \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t &= \langle \hat{a}_i \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c + \langle \hat{a}_i \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \\ &\quad - \langle \hat{a}_i \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \rangle_t^c. \end{aligned} \quad (16)$$

Through the canonical commutation relations (4) it is not difficult to see that the correlations must meet the following symmetry conditions:

$$\begin{aligned} \langle \dots \hat{a}_i \hat{a}_{j'}^\dagger \dots \rangle_t^c &= -\langle \dots \hat{a}_{j'}^\dagger \hat{a}_i \dots \rangle_t^c, \\ \langle \dots \hat{a}_i \hat{a}_{i_2} \dots \rangle_t^c &= -\langle \dots \hat{a}_{i_2} \hat{a}_i \dots \rangle_t^c, \\ \langle \dots \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \dots \rangle_t^c &= -\langle \dots \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \dots \rangle_t^c. \end{aligned} \quad (17)$$

Thus, by means of (15) the polarization components (11) can be expressed via correlations in a trivial way:

$$\mathfrak{P}_k^i = \langle \hat{a}_i \hat{a}_{j'}^\dagger \rangle_t^c, \quad \mathbf{i}' = (c, \mathbf{k}), \quad \mathbf{j}' = (v, \mathbf{k}). \quad (18)$$

In addition, there is another important quantity worth rewriting in terms of correlations—particle density operators:

$$\begin{aligned} \mathbf{n}_{\lambda, \mathbf{k}}^i &= \langle \hat{n}_{\lambda, \mathbf{k}} \rangle_t^c = \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t^c, \quad \mathbf{i}' = \mathbf{j}' = (\lambda, \mathbf{k}), \\ \langle \hat{a}_i \hat{a}_{j'}^\dagger \rangle_t^c &= 1 - \mathbf{n}_{\lambda, \mathbf{k}}^i. \end{aligned} \quad (19)$$

Further, we will see that taking into account some other physical assumptions only these two functions enter the EOM. Also, within the present study we do not consider the correlations higher than three-particle ones. In such situation it looks very reasonable to introduce specific variables. For two- and three-particle correlations they are as follows:

$$\mathfrak{D}_{i_1, i_2, j_2, j_1}^i = \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c, \quad (20)$$

$$\mathfrak{I}_{i_1, i_2, i_3, j_3, j_2, j_1}^i = \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{i_3} \hat{a}_{j_3}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c, \quad (21)$$

where all \mathbf{i}' and \mathbf{j}' are compound indices. Here one should make a remark regarding the features of notation. The expressions $\langle \hat{a}_1 \dots \hat{a}_N \hat{a}_N^\dagger \dots \hat{a}_1^\dagger \rangle_t$ and $(2N)_t$ are used for the average of the $2N$ -operator product, while $\langle \hat{a}_1 \dots \hat{a}_N \hat{a}_N^\dagger \dots \hat{a}_1^\dagger \rangle_t^c$ and $\langle N \rangle_t^c$ for correlations of N th order.

Having obtained an idea about correlations, let us finally figure out what is the benefit to work with them instead of expectation values. The understanding could be best achieved by comparing the structures of the EOM in both cases. Based on the interaction form (3), following the notation in [37]

schematically for $2N$ -operator expectation values the differential equation can be written as follows:

$$\hbar \frac{d}{dt} \langle 2N \rangle_t = \tilde{T}_N [\langle 2N \rangle_t] + \tilde{V}_{2,N} [\langle 2N + 2 \rangle_t], \quad (22)$$

while for N -particle correlations the corresponding EOM has the following form:

$$\begin{aligned} \hbar \frac{d}{dt} \langle N \rangle_t^c &= T_N [\langle N \rangle_t^c] + V_{2,N} [\langle N + 1 \rangle_t^c] \\ &\quad + V_{1,N} [\langle N + 1 \rangle_{(N)_t^c, (N-1)_t^c, \dots, (1)_t^c}]. \end{aligned} \quad (23)$$

From (22) one can see that in order to obtain a closed system one has to omit the term $\tilde{V}_{2,N}$, but in this case we are totally losing the information about contributions into dynamics from the interaction. In the case of correlations, however, the presence of $V_{1,N}$ which contains correlation of order no higher than N allows us to construct a closed system of differential equations without losing the interaction information within the given order of approximation. Thus, in order to obtain a closed system of the EOM within an N -particle correlation approximation, we shall neglect all correlations of order $N + 1$ and higher. In the next section in terms of this schematic equation we describe all the approximations for which analytical and partly numerical results with some simplification are presented in this paper.

B. Series of approximations

1. Free system dynamics

First, for the completeness of the study we reproduce the expression for free-particle polarization. The corresponding schematic equation by means of (23) reads

$$\hbar \frac{d}{dt} \langle 1 \rangle_t^c = T_1 [\langle 1 \rangle_t^c], \quad (24)$$

where we omit all the contributions connected with Coulomb interaction.

2. One-particle dynamics

Following the adopted strategy, within the one-particle correlation approximation the EOM looks like

$$\hbar \frac{d}{dt} \langle 1 \rangle_t^c = T_1 [\langle 1 \rangle_t^c] + V_{1,1} [\langle 2 \rangle_{(1)_t^c}]. \quad (25)$$

The notation $\langle 2 \rangle_{(1)_t^c}$ means that two-particle correlations enter the corresponding equation only via the product of one-particle correlations. Further, based on this expression, the well-known semiconductor Bloch equation is restored.

3. Two-particle dynamics via one-particle correlations

As the next step we include into consideration two-particle correlation dynamics; however, within this approximation only one-particle contributions enter the corresponding EOM. These corrections can be associated with scattering processes. Let us note that the EOM for one-particle correlations is already exact. Recapitulating what was said above we come

to the following system:

$$\begin{aligned}\hbar \frac{d}{dt} \langle 1 \rangle_t^c &= T_1[\langle 1 \rangle_t^c] + V_{1,1}[\langle 2 \rangle_{(1)_t^c}] + V_{2,1}[\langle 2 \rangle_t^c], \\ \hbar \frac{d}{dt} \langle 2 \rangle_t^c &= K[T_2[\langle 2 \rangle_t^c]] + V_{1,2}[\langle 3 \rangle_{(1)_t^c}],\end{aligned}\quad (26)$$

where operator K discards from T_2 all the explicitly field-dependent parts. In terms of two-particle correlations it formally can be expressed as

$$K[\langle [A, H_{el,k}] \rangle_t + \langle [A, H_I] \rangle_t] = \langle [A, H_{el,k}] \rangle_t. \quad (27)$$

This operation allows us to select from the right-hand side of the second equation in (26) only the one pure two-particle term which totally coincides with those from the left-hand side.

4. Two-particle dynamics

Here, in addition to the previous case, we include into the second line of (26) two-particle correlation terms themselves. Thus, the corresponding system reads

$$\begin{aligned}\hbar \frac{d}{dt} \langle 1 \rangle_t^c &= T_1[\langle 1 \rangle_t^c] + V_{1,1}[\langle 2 \rangle_{(1)_t^c}] + V_{2,1}[\langle 2 \rangle_t^c], \\ \hbar \frac{d}{dt} \langle 2 \rangle_t^c &= T_2[\langle 2 \rangle_t^c] + V_{1,2}[\langle 3 \rangle_{(2)_t^c, (1)_t^c}].\end{aligned}\quad (28)$$

The second line of system (28) as will be further demonstrated has a very different form for different types of two-particle correlations. This feature stems from the desire to omit all the quadratically and higher field-dependent contributions into polarization dynamics. One of the effects appearing with including into consideration the pure two-particle correlations is connected with screening of the Coulomb interaction which affects the one-particle correlations dynamics by means of a coupled system of differential equations.

5. Three-particle dynamics via one- and two-particle correlations

In this approximation the three-particle terms are taken into account. First, however, we omit pure three-particle contributions. Here, within the linear in field approximation the EOMs for one- and two-particle correlations are exact. Thus, the corresponding system reads

$$\begin{aligned}\hbar \frac{d}{dt} \langle 1 \rangle_t^c &= T_1[\langle 1 \rangle_t^c] + V_{1,1}[\langle 2 \rangle_{(1)_t^c}] + V_{2,1}[\langle 2 \rangle_t^c], \\ \hbar \frac{d}{dt} \langle 2 \rangle_t^c &= T_2[\langle 2 \rangle_t^c] + V_{1,2}[\langle 3 \rangle_{(2)_t^c, (1)_t^c}] + V_{2,2}[\langle 3 \rangle_t^c], \\ \hbar \frac{d}{dt} \langle 3 \rangle_t^c &= K[T_3[\langle 3 \rangle_t^c]] + V'_{1,3}[\langle 4 \rangle_{(2)_t^c, (1)_t^c}].\end{aligned}\quad (29)$$

The primed function $V'_{1,3}$ means that only terms without momentum summation are considered. This will be discussed later.

Within this paper we limit ourselves only by analyzing three-particle contributions. This is motivated by the will to obtain trion-like behavior of the absorption spectrum. Moreover, due to the noticeable numerical complexity we deviate from the presented set of approximations at the last step by introducing some simplifications which will be discussed further.

IV. RESULTS

A. Analytics

Before we proceed with analytics and numerics, let us make some assumptions, which allows to dramatically decrease the complexity of further computations. From now, following the problem statement in [37] we suppose that the analyzed systems are excited only by a homogeneous electric field with polarization lying in the sample plane. This results in coincidence of total momenta of annihilation and creation operators in expectation values. In terms of one-particle correlations this requirement reads as

$$\langle \hat{a}_i \hat{a}_{j'}^\dagger \rangle_t^c = \delta_{\mathbf{k}, \mathbf{k}'} \langle \hat{a}_i \hat{a}_{j'}^\dagger \rangle_t^c, \quad (30)$$

with compound indices $\mathbf{i}' = (\lambda, \mathbf{k})$ and $\mathbf{j}' = (\lambda', \mathbf{k}')$. This simplification leads to the fact that among all one-particle correlations only $\mathfrak{P}(\mathbf{k}, t)$ and $n(\lambda, \mathbf{k}, t)$ survive in the EOM. In the general case, the homogeneity condition is expressed as follows:

$$\begin{aligned}\langle \hat{a}_{i'_1} \dots \hat{a}_{i'_n} \hat{a}_{j'_n}^\dagger \dots \hat{a}_{j'_1}^\dagger \rangle_t^c \\ = \delta_{\mathbf{k}_1 + \dots + \mathbf{k}_n, \mathbf{k}'_1 + \dots + \mathbf{k}'_n} \langle \hat{a}_{i'_1} \dots \hat{a}_{i'_n} \hat{a}_{j'_n}^\dagger \dots \hat{a}_{j'_1}^\dagger \rangle_t^c,\end{aligned}\quad (31)$$

with index structure similar to (30). In fact, this assumption allows one to reduce a dimension of all integrals appearing within calculations. Within the body of the paper we present only the final expressions for low-order approximations. Due to the declared interest in description of optical spectra, the results will be presented for the \mathbf{k} component of susceptibility $\chi(\omega)$ entering the following relation:

$$\tilde{\mathfrak{P}}_{\mathbf{k}}^\omega = \chi(\mathbf{k}, \omega) \mathcal{E}(\omega). \quad (32)$$

It should be noted that electric susceptibility $\chi(\omega)$ is one of the most calculated quantities due to the fact that it contains a lot of information about the optical properties of materials including oscillator strength, absorption, refractive index, etc. The TMDs are not an exception; there are plenty of works where the susceptibility was analyzed by means of different theoretical approaches (see, e.g., Refs. [49,50] and references therein).

In this work, due to the equilibrium system requirements, we also replace all $n'_{\lambda, \mathbf{k}}$ by their equilibrium values, i.e., Fermi-Dirac distribution functions. Moreover, from the computational point of view it is quite useful to work with holes instead of valence electrons. The corresponding relations are as follows:

$$f_{(c, \mathbf{k})} = \frac{1}{e^{\beta(E_g + \hbar^2 \mathbf{k}^2 / 2m_c - \mu_c)} + 1}, \quad (33)$$

$$f_{(v, \mathbf{k})} = \frac{1}{e^{\beta(\hbar^2 \mathbf{k}^2 / 2m_v - \mu_v)} + 1}, \quad (34)$$

$$f_{(h, \mathbf{k})} = \frac{1}{e^{\beta(\hbar^2 \mathbf{k}^2 / 2m_h - \mu_h)} + 1}, \quad (35)$$

$$f_{(v, \mathbf{k})} = 1 - f_{(h, \mathbf{k})}, \quad (36)$$

where E_g is the band gap energy, $\beta = 1/kT$, m_h and m_v are the hole and valence electron masses ($m_h = -m_v$, with $m_h > 0$), while μ_h and μ_v are the chemical potentials of

holes and valence electrons, respectively ($\mu_h = -\mu_v$), and m_c is the conduction band electron mass. Also, we use further $f_{(e,k)} \equiv f_{(c,k)}$ and $m_e \equiv m_c$. All the details of calculation can be found in Appendix C.

1. Free system dynamics

Within this approximation the cluster expansion technique is unnecessary. We set the potential equal to zero and derive from Eq. (12) the following expression:

$$\chi_I(\mathbf{k}, \omega) = -\frac{d_{cv}[1 - f_{(e,k)} - f_{(h,k)}]}{\hbar[\omega + i\delta - (\varepsilon_{(e,k)} + \varepsilon_{(h,k)})]}, \quad (37)$$

where $\varepsilon_{(e,k)}$ and $\varepsilon_{(h,k)}$ are one-particle energies, defined in (C18). This result coincides with the well-known formula for susceptibility of noninteracting systems.

2. One-particle dynamics and semiconductor Bloch equation

From (25) by means of the cluster expansion (14) we find

$$\chi_{II}(\mathbf{k}, \omega) = \Gamma_2(\mathbf{k})\chi_I^R(\mathbf{k}, \omega), \quad (38)$$

$$\begin{aligned} \chi(\mathbf{k}, \omega) &= \chi_I^R(\mathbf{k}, \omega) \left[1 + \frac{1}{d_{cv}} \sum_{q \neq k} V_{k-q} \chi(\mathbf{q}, \omega) \right] + \frac{1}{\mathcal{E}(\omega)\hbar[\omega + i\delta - (\varepsilon_{(c,k)} - \varepsilon_{(v,k)})]} \\ &\times \sum_{k'_2, q' \neq 0} V_{q'} [\mathfrak{D}_{(c,k-q'), (c,k'_2), (c,k'_2-q'), (v,k)}^\omega + \mathfrak{D}_{(c,k-q'), (v,k'_2), (v,k'_2-q'), (v,k)}^\omega \\ &- \mathfrak{D}_{(c,k), (v,k'_2), (v,k'_2-q'), (v,k+q')}^\omega - \mathfrak{D}_{(c,k), (c,k'_2), (c,k'_2-q'), (v,k+q')}^\omega]. \end{aligned} \quad (41)$$

Within all the further calculations the structure of this equation remains the same. Only the functions \mathfrak{D} will be calculated within the different approximation orders. In order to get an idea about the structure of functions \mathfrak{D} , from the second line of the system (26) within the assumptions adopted in this paper we find the following equation for one of the terms in (41):

$$\mathfrak{D}_{(c,k-q'), (c,k'_2), (c,k'_2-q'), (v,k)}^{\omega, III} = F_{\mathfrak{D},1}(\chi_{II}, \mathbf{k} - \mathbf{q}', \mathbf{k}'_2, \mathbf{k}'_2 - \mathbf{q}', \mathbf{k}, \omega, \{cccv\}), \quad \text{with function } F_{\mathfrak{D},1} \text{ defined as} \quad (42)$$

$$\begin{aligned} F_{\mathfrak{D},1}(\chi, \mathbf{k} - \mathbf{q}', \mathbf{k}'_2, \mathbf{k}'_2 - \mathbf{q}', \mathbf{k}, \omega, \{cccv\}) &= \frac{\mathcal{E}(\omega)}{\hbar[\omega + i\delta - [\varepsilon_{(c,k-q')} + \varepsilon_{(c,k'_2)} - \varepsilon_{(c,k'_2-q')} - \varepsilon_{(v,k)}]]} \\ &\times [\chi(\mathbf{k}, \omega)V_{k-k'_2}[-f_{(c,k'_2)}f_{(c,k-q')} + f_{(c,k'_2-q')}[-1 + f_{(c,k'_2)} + f_{(c,k-q')}] \\ &+ \chi(\mathbf{k}'_2, \omega)V_{k-k'_2}[f_{(c,k-q')}[1 - f_{(c,k'_2-q')} - f_{(v,k)}] + f_{(c,k'_2-q')}f_{(v,k)}] \\ &+ \chi(\mathbf{k}, \omega)V_{q'}[f_{(c,k'_2)}f_{(c,k-q')} + f_{(c,k'_2-q')}[1 - f_{(c,k'_2)} - f_{(c,k-q')}] \\ &+ \chi(\mathbf{k} - \mathbf{q}', \omega)V_{q'}[f_{(c,k'_2)}[-1 + f_{(c,k'_2-q')} + f_{(v,k)}] - f_{(c,k'_2-q')}f_{(v,k)}]]. \end{aligned} \quad (43)$$

As can be seen, we introduce the superscripts for two-particle correlations: $\mathfrak{D}^{\omega,3}$. This is dictated by the needs of numerical calculations. For the approximate solution of the emergent system of algebraic equations we develop an iterative procedure. Let us briefly describe it. All the functions which we introduce here can be found in Appendix C 3. As the first basic step we take the solution obtained by means of Eq. (38):

$$\chi_{II}(\mathbf{k}, \omega) = F_{\chi,1}(\chi_{II}, \mathbf{k}, \omega). \quad (44)$$

As was mentioned above, this equation can be solved by the matrix inversion approach. On the basis of this initial point,

where functions Γ_{II} and χ_I^R are as follows:

$$\chi_I^R(\mathbf{k}, \omega) = -\frac{d_{cv}[1 - f_{(e,k)} - f_{(h,k)}]}{\hbar[\omega + i\delta - (\varepsilon_{(e,k)} + \varepsilon_{(h,k)})]}, \quad (39)$$

$$\Gamma_{II}(\mathbf{k}) = 1 + \frac{1}{d_{cv}} \sum_{q \neq k} \chi_I^R(\mathbf{q}, \omega)V_{k-q}\Gamma_{II}(\mathbf{q}). \quad (40)$$

The superscript R denotes that the energies in the denominator of (39) contain renormalized energies in contrast to (37). It can also be noted that $\Gamma_{II}(\mathbf{k})$ coincides with the generalized Rabi frequency up to a factor $d_{cv}\mathcal{E}/\hbar$. The details of calculation are presented in Appendix C 2. This result coincides with answer obtained by means of the well-known semiconductor Bloch equation [38].

3. Multiparticle analysis

Starting from the approximation (26) the equation for dynamics of one-particle correlations is already exact. However, it contains the terms which we do not know explicitly. Fortunately, for these contributions some equations can be derived.

For the susceptibility component from the first line of (26) one can obtain by means of (C38) and (21) the following expression:

one can iteratively find further approximations presented in the previous section by means of the following expressions. For $N \in \{III, IV\}$ we have

$$\begin{aligned} \mathfrak{D}_{(\lambda_1, \mathbf{p}_1), (\lambda_2, \mathbf{p}_2), (\lambda_3, \mathbf{p}_3), (\lambda_4, \mathbf{p}_4)}^{\omega, N} &= F_{\mathfrak{D},1}(\chi_{II}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1\lambda_2\lambda_3\lambda_4\}) \\ &+ F_{\mathfrak{D},2}(\mathfrak{D}^{\omega, N-1}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1\lambda_2\lambda_3\lambda_4\}), \\ \chi_N(\mathbf{k}, \omega) &= F_{\chi,1}(\chi_N, \mathbf{k}, \omega) + F_{\chi,2}(\mathfrak{D}^{\omega, N}, \mathbf{k}, \omega), \end{aligned} \quad (45)$$

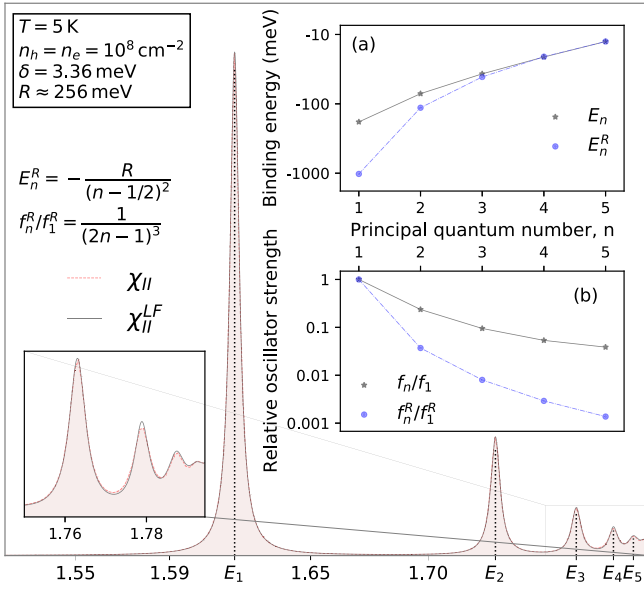


FIG. 2. The behavior of absorption spectrum in the low-density regime ($n_e = n_h = 10^8 \text{ cm}^{-2}$). Inset (a) demonstrates the deviation of our results (E_n) for exciton binding energies from the hydrogenic series of an ideal 2d system (E_n^R). The Rydberg constant R is calculated under the following condition: $E_5 = E_5^R$. Inset (b) demonstrates the deviation of our results (f_n/f_1) for relative oscillator strength from the 2d hydrogenic model of excitons (f_n^R/f_1^R).

where by definition we consider $\mathcal{D}^{\omega,I} \equiv \mathcal{D}^{\omega,II} \equiv 0$. Having obtained the function $\mathcal{D}^{\omega,N}$ one can calculate the modified solution for $\chi_N(\mathbf{k}, \omega)$ again by means of the matrix inversion approach. The analysis of $N = V$ is isolated, due to the needs of applying some simplification. Owing to the computational requirements, the corresponding system of equations for $N = V$ reads

$$\begin{aligned} & \mathcal{T}_{(\lambda_1, p_1), (\lambda_2, p_2), (\lambda_3, p_3), (\lambda_4, p_4)}^{\omega, V} \\ &= F_{\mathcal{T}, I}(\chi_{II}, \mathcal{D}^{\omega, III}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}), \\ & \mathcal{D}_{(\lambda_1, p_1), (\lambda_2, p_2), (\lambda_3, p_3), (\lambda_4, p_4)}^{\omega, V} \\ &= F_{\mathcal{D}, III}(\mathcal{T}^{\omega, V}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}), \\ & \chi_V(\mathbf{k}, \omega) \\ &= F_{\chi, 1}(\chi_V, \mathbf{k}, \omega) + F_{\chi, 2}(\mathcal{D}^{\omega, V}, \mathbf{k}, \omega). \end{aligned} \quad (46)$$

As was said previously, all the presented functionals F can be found in Appendix C3 and C4. Also, they are put into separate Mathematica files, which can be found in the Supplemental Material [51].

B. Numerics and discussion

In this section we obtain and compare the absorption spectra for different approximations and physical parameter values (see Fig. 2). Due to the relatively low computational cost of obtaining $\chi_{II}(\mathbf{k}, \omega)$, we present it for a wide range of carrier densities and temperatures. We note however that since we did not account for the electron-phonon interaction, the temperature here only affects the smearing of the Fermi surface. For some relevant values of temperature and concen-

tration combinations we obtain higher-order corrections up to $\chi_V(\mathbf{k}, \omega)$, where as expected the trion-like resonance manifest itself. The detailed description of the numerical computations can be found in Appendix D. Let us present here only the values of some physical parameters. For the average dielectric constant and effective screening length we choose the following reasonable numbers: $\epsilon_0 = 2$ and $r_0 = 5 \text{ nm}$. As for the effective masses of electrons and holes we stopped on the typical values: $m_e = 0.4m_0$ and $m_h = 0.6m_0$, where m_0 is the electron rest mass. The band gap energy E_g is set to 1.8 eV. All the other parameters which are meaningless from the physical point of view and affect only the efficiency of the calculation procedure are defined and discussed in Appendix D.

In order to assess the correctness of the analytical calculations presented in the previous section, we analyze a number of dependencies. First, the dependence of resonance position on carrier concentrations is analyzed at fixed temperatures. Next, we study the excitonic resonance behavior relative to the doped carrier concentration. Also, some additional dependencies which are usually obtained via experiments will be presented.

1. Carrier concentration variation

In this case we fix the temperature and vary the densities. We limit ourselves to temperatures 5 K, 100 K, and 273 K. The corresponding dependencies are presented in Fig. 3. It should be noted that high-temperature behavior cannot be described without taking into account the phonons; therefore in the cases of 273 K and 100 K the pictures cannot be perceived as genuine ones. Also, we considered separately the low-density regime when the electron-hole pair concentration is equal to 10^8 cm^{-2} Fig. 2. We compare the calculated binding energy series with the analytical formula for exciton energies of an ideal 2d system: $-R/(n-1/2)^2$. Assuming that for large principal quantum numbers (in our case $n = 5$) both dependencies have to coincide, we extract the Rydberg constant: $R = 256 \text{ meV}$. It should be noted that the estimate obtained by means of analytical expression is much greater: $e^4 m_r / 2\epsilon_0 \hbar^2 \approx 816 \text{ meV}$. It is seen that our results deviate from this simple model dependence which is generally accepted for TMDs [1,4,6]. For the case of 5 K, we also obtain the drift of $1s$ exciton peak position with growth of electron-hole pair concentration within the χ_{II} and χ_{III} approximations. The corresponding trend is depicted in Fig. 4. The extracted estimate of slope ratio ($\approx 0.0026 \text{ meV } \mu\text{m}^2$) is of the same order of magnitude as the number recently obtained within the experiment [52] on TMD MLs. This number allows one to extract the exciton Bohr radius by means of the following relation: $dE_1/dn_r = 2.07E_1 a_B^2$ [53]. It leads to the following estimate: $a_B = 2.6 \text{ nm}$. Also, we observe the onset of the Mott transition at the concentrations with $n_r \approx 10, 170, 400$ for 5 K, 100 K, and 273 K, respectively. At these concentrations the absorption peaks associated with the $1s$ exciton disappear and the system is characterized by negative absorption coefficients. Moreover, at low temperature $T = 5 \text{ K}$ we observe the negative absorption region in the vicinity of the second exciton state. This is due to the fact that since we disregard the electron-phonon interaction, the excited exciton states do not relax to

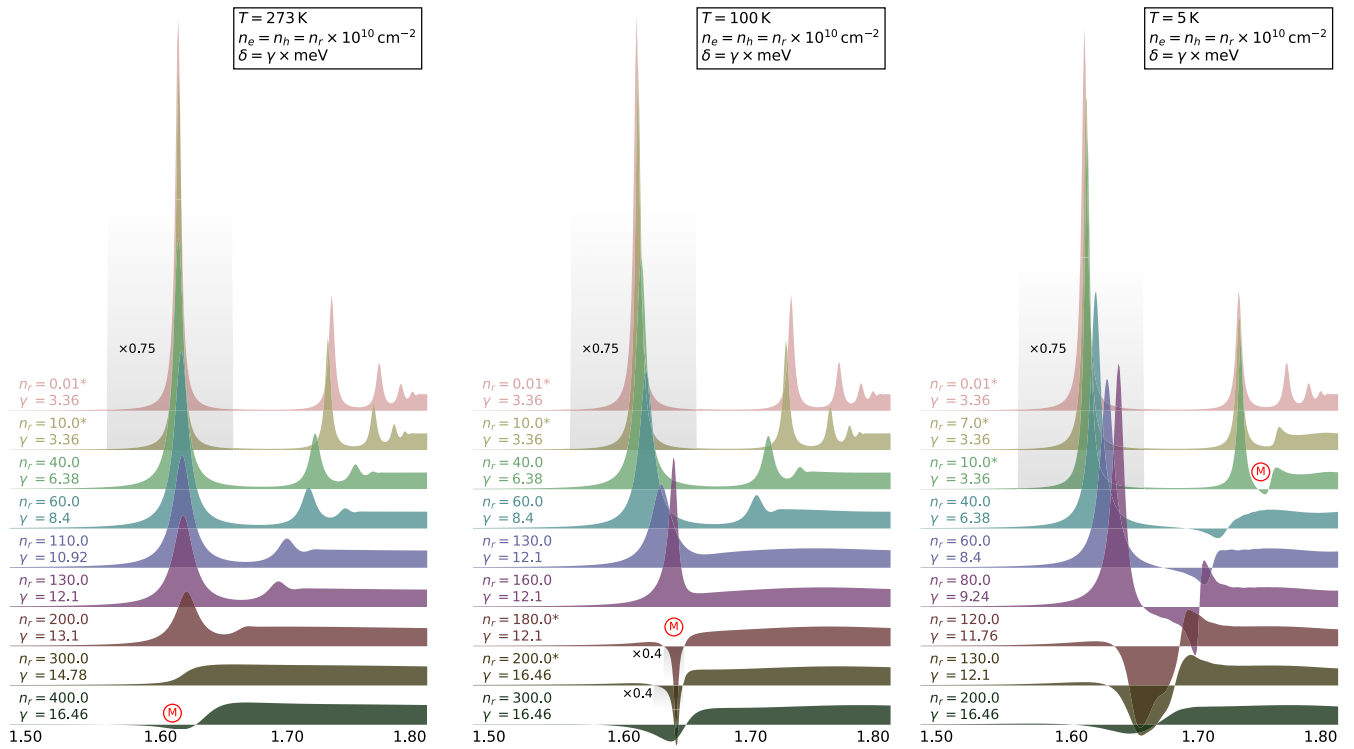


FIG. 3. Change of absorption spectra behavior with increasing of exciton concentration in the case of three different temperature values: 273 K, 100 K, 5 K. The circled letter M indicates the vicinity of the Mott transition. The star sign as a superscript for different n_r values means that the corresponding spectrum has rescaled regions. The shaded areas demonstrate that the corresponding dependence has a rescaled region.

the ground ones. We note also that since we only consider a single valley in our model, the results cannot be directly compared to the experimentally observed exciton shifts in TMDs since no contribution of the intervalley scattering can be accounted for.

In Fig. 5 we plot the dependence of the difference between exciton shifts obtained within the χ_{II} and χ_{III} approximations.

2. Doping level variation

In this section as previously we choose the same temperature values set but fix the number of hole concentration at

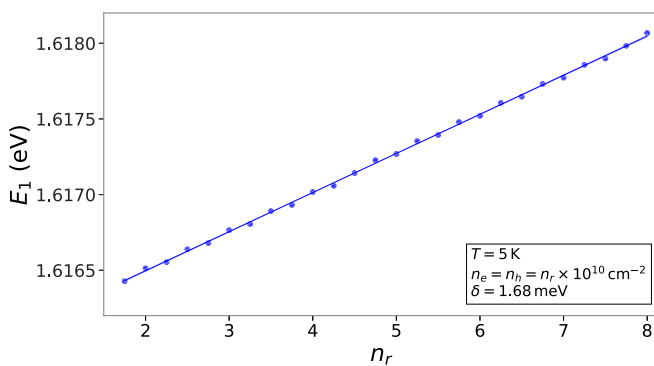


FIG. 4. The dependence of $1s$ ($n = 1$) excitonic peak position on the electron-hole concentration within χ_{II} approximation. The linear regression gives the following result for the slope ratio $|dE_1/dn_e| = 0.0026 \text{ meV } \mu\text{m}^2$.

value 10^{10} cm^{-2} . It is worth noting here that the picture would change only slightly if we set the hole concentration to zero. In particular, the peak position would remain almost unchanged. The corresponding dependencies are depicted in Fig. 6. For zero-valued hole concentration we calculate the behavior of $1s$ excitonic peak position with growth of electron density which is presented in Fig. 7. The value of the slope ratio extracted from this dependence is equal to $\approx 1.0 \mu\text{eV } \mu\text{m}^2$. The corresponding dependence of difference between $1s$ excitonic peak position obtained within different approximations on the electron-hole concentration is also presented in Fig. 9. We

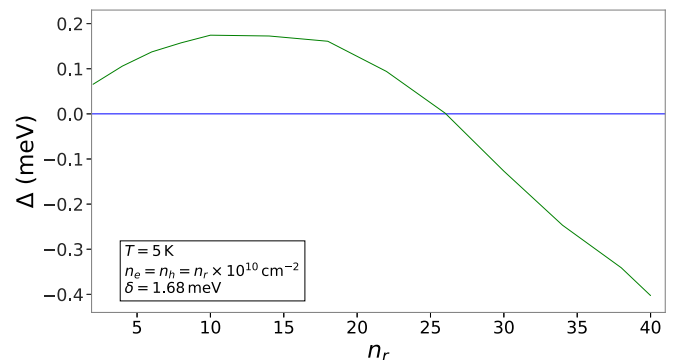


FIG. 5. The dependence of difference between $1s$ excitonic peak position obtained within χ_{II} and χ_{III} approximations on the electron-hole concentration ($\Delta = E_1^{\chi_{II}} - E_1^{\chi_{III}}$).

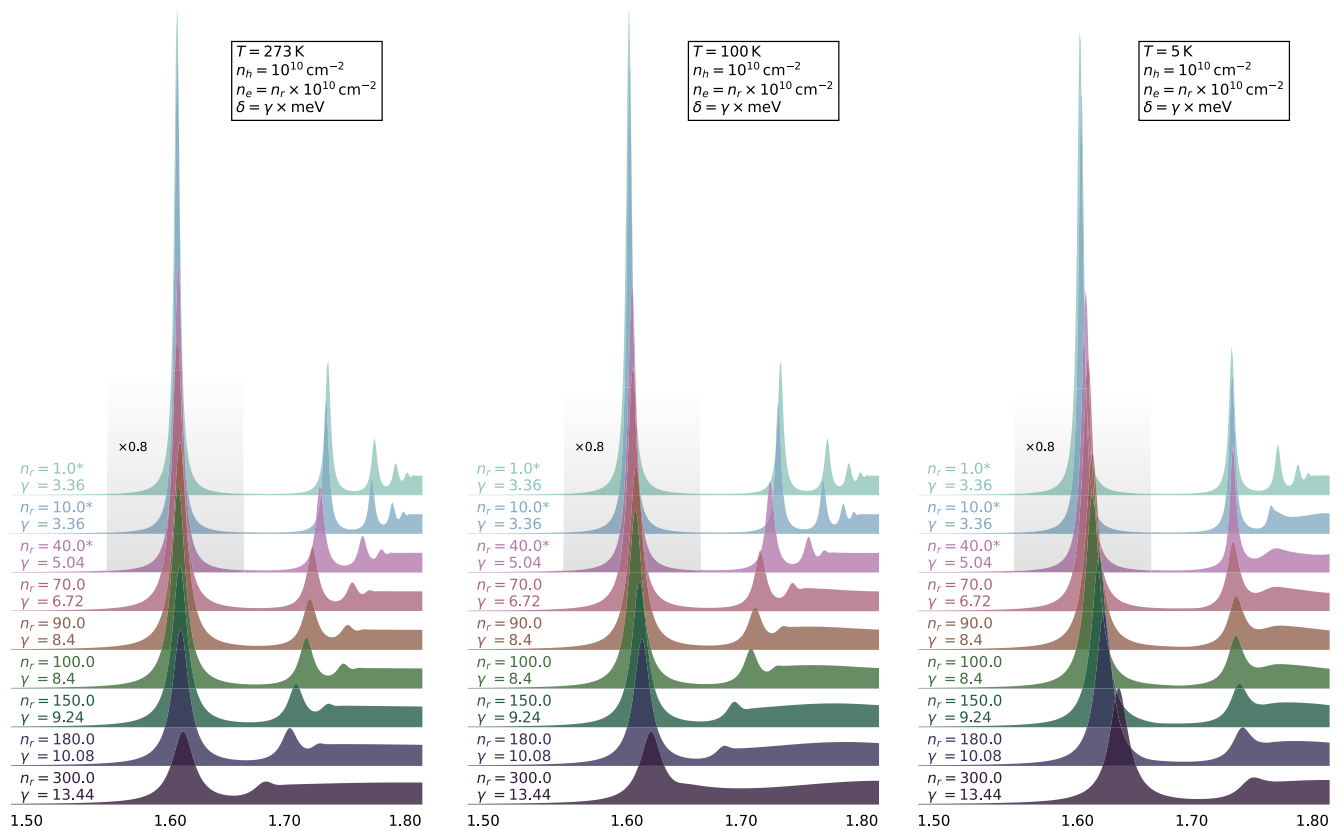


FIG. 6. Change of absorption spectra behavior with increasing of electron concentration when hole density is zero. The dependencies are presented for three different temperature values: 237 K, 100 K, 5 K.

also obtain the dependence of the normalized peak height on electron density which is presented in Fig. 8.

In the case when the electron concentration exceeds the hole one it is reasonable to perform the search of trionic states. For such analysis we choose the following set of calculation parameters: $T = 5$ K, $n_h = 10^{10} \text{ cm}^{-2}$, $n_e = 40n_h$, $\delta = 6.72$ meV. The results of the corresponding computations are presented in Fig. 10. The moderate scattering of calculated points from the expected Lorenz-like behavior of peaks is caused by the integration accuracy of corrections.

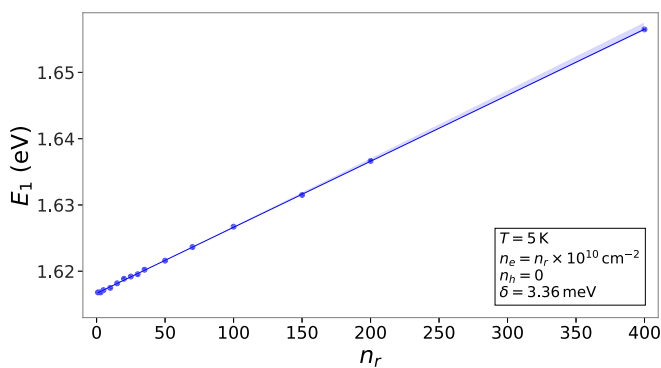


FIG. 7. The dependence of $1s$ excitonic peak position on the electron concentration when hole density is equated to zero. The linear regression gives the following result for the slope ratio: $|dE_1/dn_e| = 0.000996 \text{ meV } \mu\text{m}^2$.

The detailed description of the numerical procedure is presented in Appendix D2. The fitting of obtained results by means of Lorentzian functions allows us to extract the numerical estimate for trion binding energy. It turns out to be about 6 meV which is at least twice smaller than the experimentally observed values [54,55]. This discrepancy is mainly due to the abundance of the multivalley structure of TMDs in our model, since it is known that the fundamental trion state corresponds to the two electrons filling different valleys.

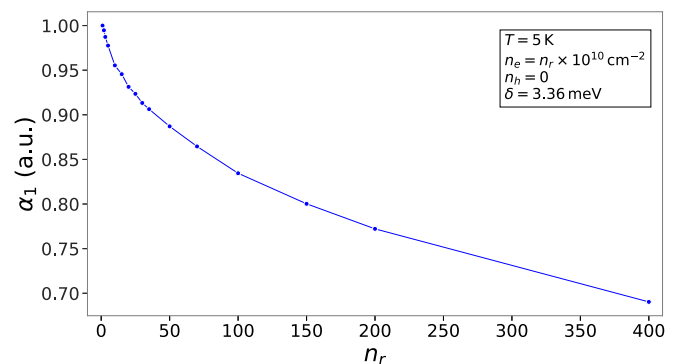


FIG. 8. The behavior of peak height for $1s$ excitonic state. The absorption is normalized to unity, which, however, allows us to understand the relative change.

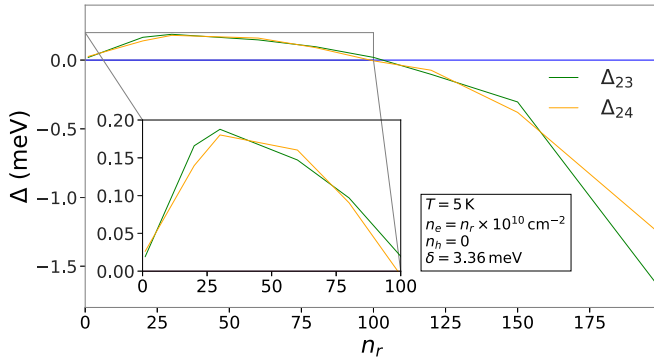


FIG. 9. The dependence of difference between $1s$ excitonic peak position obtained within χ_{II} , χ_{III} , and χ_{IV} approximations on the electron-hole concentration ($\Delta_{23} = E_1^{\chi_{III}} - E_1^{\chi_{IV}}$, $\Delta_{24} = E_1^{\chi_{III}} - E_1^{\chi_{IV}}$).

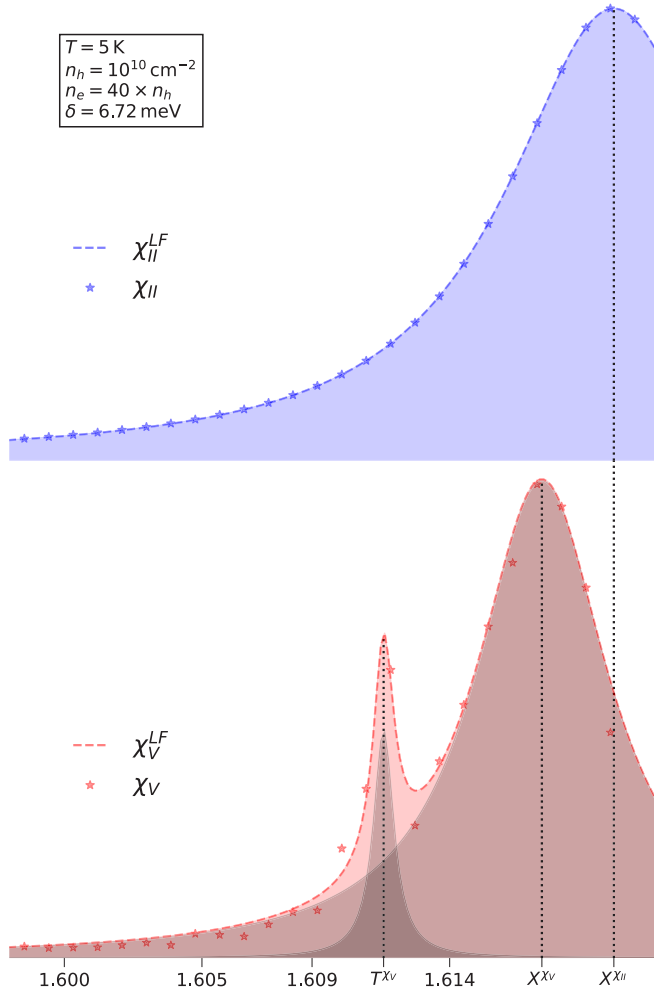


FIG. 10. The behavior of absorption spectra calculated by means of χ_{II} and χ_V approximations. The “LF” superscript denotes that the corresponding dependence is constructed by means of Lorentzian function(s) which fit the calculated points. The last in turn are denoted with the \star symbol. The quantities $X^{\chi_{II}}$, X^{χ_V} , and T^{χ_V} are equal to 1.6199(7) eV, 1.6174(7) eV, and 1.6116(6) eV, respectively.

V. CONCLUSION

To conclude, we have extended the cluster expansion technique to account for the higher-order correlations and applied it to model the optical absorption of the TMD monolayer. Our account of the three-particle correlations allowed us to model the absorption peaks associated with the trion quasiparticles. The developed technique allows us to directly model experimentally accessible absorption spectra. The main advantage of the proposed formalism is that despite the computational complexity, it allows us to address the regime of large electron-hole densities and large doping, when the Fermi energy becomes comparable to the exciton and trion binding energies and, ultimately, the exciton Mott transition.

The natural development of the presented formalism would be to include the electron-phonon interaction as well as spin and valley degrees of freedom. The cluster expansion technique proved to be a powerful tool to model the absorption spectra mediated by the strong many-body correlations present in TMD monolayers and heterostructures.

ACKNOWLEDGMENTS

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APPENDIX A: INFORMATION ABOUT SUPPLEMENTAL MATERIAL

All the functionals presented in this paper in terms of correlations can be found in the Mathematica file “convoluted_expressions_in_terms_of_arb_mom.nb” in the Supplemental Material [51].

APPENDIX B: CONNECTION BETWEEN NOTATIONS

Here we present the connection formulas between two types of notation in (2) and (3). For the model considered in this work these expressions are as follows:

$$\begin{aligned}
 \hbar h_{ij}^{\text{ext}}(t) &= -\mathcal{E}(t) d_{cv} \sum_{\mathbf{k}} [\delta_{j,(v,\mathbf{k})} \delta_{i,(c,\mathbf{k})} + \delta_{j,(c,\mathbf{k})} \delta_{i,(v,\mathbf{k})}], \\
 v_{i_1, i_2, j_1, j_2} &= \frac{1}{\hbar} \sum_{\substack{\mathbf{k}_1, \mathbf{k}_2 \\ \mathbf{q} \neq \mathbf{0}}} 2V_q [\Re \text{tron}(i_1, i_2, j_2, j_1) - \Re \text{tron}(i_2, i_1, j_2, j_1) \\
 &\quad + \Re \text{tron}(i_2, i_1, j_1, j_2) - \Re \text{tron}(i_1, i_2, j_1, j_2)], \\
 \Re \text{tron}(i_1, i_2, j_2, j_1) &= \frac{1}{4} [\delta_{(c, \mathbf{k}_1 + \mathbf{q}), i_1} \delta_{(c, \mathbf{k}_2 - \mathbf{q}), i_2} \delta_{(c, \mathbf{k}_2), j_2} \delta_{(c, \mathbf{k}_1), j_1} \\
 &\quad + \delta_{(v, \mathbf{k}_1 + \mathbf{q}), i_1} \delta_{(v, \mathbf{k}_2 - \mathbf{q}), i_2} \delta_{(v, \mathbf{k}_2), j_2} \delta_{(v, \mathbf{k}_1), j_1} \\
 &\quad + \delta_{(c, \mathbf{k}_1 + \mathbf{q}), i_1} \delta_{(v, \mathbf{k}_2 - \mathbf{q}), i_2} \delta_{(v, \mathbf{k}_2), j_2} \delta_{(c, \mathbf{k}_1), j_1} \\
 &\quad + \delta_{(v, \mathbf{k}_1 + \mathbf{q}), i_1} \delta_{(c, \mathbf{k}_2 - \mathbf{q}), i_2} \delta_{(c, \mathbf{k}_2), j_2} \delta_{(v, \mathbf{k}_1), j_1}].
 \end{aligned} \tag{B1}$$

$$\begin{aligned}
 (\times 2) \quad & [\hat{D} + \hat{E}] \text{---} \bullet \text{---} \bullet = \text{---} \bullet \text{---} \bullet \text{---} \text{E} \text{---} \bullet \text{---} \bullet \\
 & = i\hbar[\varepsilon_{i'} - \varepsilon_{j'}]\langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c. \quad (C2)
 \end{aligned}$$

FIG. 11. The differential equation on one-particle correlations within free-system approximation ($N = I$).

These artificially long expressions are caused by the symmetrization conditions (5).

APPENDIX C: EOMs: SERIES OF APPROXIMATIONS

Because we are interested in the frequency spectrum, we introduce the Fourier transform and work further with algebraic equations instead of differential ones. Within this paper for the Fourier transform we accept the following definition:

$$\begin{aligned}
 \tilde{\mathfrak{P}}(\mathbf{k}, \omega) &= \int_{-\infty}^{\infty} dt \tilde{\mathfrak{P}}(\mathbf{k}, t) \exp(i\omega t), \\
 \tilde{\mathfrak{P}}(\mathbf{k}, t) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \tilde{\mathfrak{P}}(\mathbf{k}, \omega) \exp(-i\omega t). \quad (C1)
 \end{aligned}$$

Let us derive the general expressions which are necessary for all the approximation orders. On the basis of (12), we derive the dynamical equations on correlations. First, the commutator of $\hat{a}_{i'} \hat{a}_{j'}^\dagger$ and $H_{el,k}$ gives

$$i\langle [\hat{a}_{i'} \hat{a}_{j'}^\dagger, H_{el,k}]_t \rangle = i\hbar[\varepsilon_{i'} - \varepsilon_{j'}] \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t$$

The commutator of H_I with four- and six-operator combinations can be written by analogy:

$$\begin{aligned}
 i\langle [\hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger, H_I]_t \rangle &= i\hbar \left[\sum_j (T_{(i_1 j), (j i_2 j_2 j_1)}^4 + T_{(i_2 j), (i_1 j_2 j_2 j_1)}^4) - \sum_i (T_{(i j_2), (i_1 i_2 i j_1)}^4 + T_{(i j_1), (i_1 i_2 j_2 i)}^4) \right], \\
 i\langle [\hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{i_3} \hat{a}_{j_3}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger, H_I]_t \rangle &= i\hbar \left[\sum_j (T_{(i_1 j), (j i_2 i_3 j_3 j_2 j_1)}^6 + T_{(i_2 j), (i_1 i_3 j_3 j_2 j_1)}^6 + T_{(i_3 j), (i_1 i_2 j_3 j_2 j_1)}^6) \right. \\
 &\quad \left. - \sum_i (T_{(i j_3), (i_1 i_2 i_3 j_2 j_1)}^6 + T_{(i j_2), (i_1 i_2 i_3 j_3 i j_1)}^6 + T_{(i j_1), (i_1 i_2 i_3 j_3 j_2 i)}^6) \right], \quad (C6)
 \end{aligned}$$

where $T_{(i_1 j), (j i_2 j_2 j_1)}^4$ and $T_{(i_1 j), (j i_2 i_3 j_3 j_2 j_1)}^6$ read as

$$T_{(i_1 j), (j i_2 j_2 j_1)}^4 = h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t, \quad T_{(i_1 j), (j i_2 i_3 j_3 j_2 j_1)}^6 = h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{i_3} \hat{a}_{j_3}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t. \quad (C7)$$

Let us note, however, that these quantities, in contrast to T^2 , do not coincide with their correlation counterparts. For this purpose one should address the cluster expansion (14).

Based on the derived expressions the logic of the construction of commutators with many-operator combinations is clear. All the aforementioned relations allow one to rewrite the left-hand side of (12) for expectation values in terms of correlations,

$$\begin{aligned}
 \frac{d}{dt} \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t + \frac{1}{\hbar} i \langle [\hat{a}_{i'} \hat{a}_{j'}^\dagger, H_{el,k}]_t \rangle + \frac{1}{\hbar} i \langle [\hat{a}_{i'} \hat{a}_{j'}^\dagger, H_I]_t \rangle \\
 = \frac{d}{dt} \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c + i[\varepsilon_{i'} - \varepsilon_{j'}] \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c + i \left[\sum_j h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c - \sum_i h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i_1} \hat{a}_i^\dagger \rangle_t^c \right], \quad (C8)
 \end{aligned}$$

or in terms of schematic notation (24),

$$\hbar \frac{d}{dt} \langle 1 \rangle_t^c = \hbar \frac{d}{dt} \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c, \quad T_1[\langle 1 \rangle_t^c] = -\hbar \left[i[\varepsilon_{i'} - \varepsilon_{j'}] \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c + i \left[\sum_j h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c - \sum_i h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i_1} \hat{a}_i^\dagger \rangle_t^c \right] \right]. \quad (C9)$$

In the next sections we include the consideration of Coulomb interaction.

The similar relation is true for the operator (13):

$$\begin{aligned}
 i\langle [\hat{a}_{i_1} \dots \hat{a}_{i_n} \hat{a}_{j_n}^\dagger \dots \hat{a}_{j_1}^\dagger, H_{el,k}]_t \rangle \\
 = i\hbar[\varepsilon_{i_1} + \dots + \varepsilon_{i_n} - \varepsilon_{j_n} - \dots - \varepsilon_{j_1}] \\
 \times \langle \hat{a}_{i_1} \dots \hat{a}_{i_n} \hat{a}_{j_n}^\dagger \dots \hat{a}_{j_1}^\dagger \rangle_t. \quad (C3)
 \end{aligned}$$

The next important contribution is caused by the presence of the external field:

$$\begin{aligned}
 i\langle [\hat{a}_{i'} \hat{a}_{j'}^\dagger, H_I]_t \rangle &= i\hbar \sum_{i,j} h_{i_1 j}^{\text{ext}} [\langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \hat{a}_i^\dagger \hat{a}_j \rangle_t - \langle \hat{a}_{i_1} \hat{a}_j \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t] \\
 &= i\hbar \left[\sum_j h_{i_1 j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{j'}^\dagger \rangle_t - \sum_i h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i'} \hat{a}_i^\dagger \rangle_t \right] \\
 &= i\hbar \left[\sum_j h_{i_1 j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{j'}^\dagger \rangle_t^c - \sum_i h_{i_1 j}^{\text{ext}} \langle \hat{a}_{i'} \hat{a}_i^\dagger \rangle_t^c \right] \\
 &= i\hbar \left[\sum_j T_{(i' j), (j j')}^2 - \sum_i T_{(i j'), (i' i)}^2 \right], \quad (C4)
 \end{aligned}$$

where $T_{(i' j), (j j')}^2$ is defined as follows:

$$T_{(i' j), (j j')}^2 = h_{i_1 j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{j'}^\dagger \rangle_t = h_{i_1 j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{j'}^\dagger \rangle_t^c = T_{c, (i' j), (j j')}^2. \quad (C5)$$

1. Free system dynamics

First, let us reproduce free-system polarization. We set $V = 0$ and obtain from (C8) the following equation:

$$\frac{d}{dt} \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c + i[\varepsilon_{i'} - \varepsilon_{j'}] \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c + i \left[\sum_j h_{ij}^{\text{ext}} \langle \hat{a}_j \hat{a}_{i'}^\dagger \rangle_t^c - \sum_i h_{ij'}^{\text{ext}} \langle \hat{a}_{i'} \hat{a}_i^\dagger \rangle_t^c \right] = 0. \quad (\text{C10})$$

For the purposes of further computations it is quite instructive to introduce the diagrammatic rules and to accompany all the analytic expressions by their graphical counterparts. Moreover, the three-particle correlation dynamics will be derived only by means of the powerful method of Feynman diagrams. The detailed description of the technique with explanation of prefactor and sign choice of a diagram was presented in [36]. In this work we add only some customization. Also, we limit ourselves only by consideration of *connected* graphs which appear in equations on correlations in contrast to differential equations on expectation values where *unconnected* diagrams are also presented. The diagram elements are as follows:

$$\begin{array}{c} \begin{array}{l} i_1 \\ / \quad \backslash \\ \times \\ \backslash \quad / \\ i_2 \end{array} = \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2}, \quad \begin{array}{l} i' \\ \curvearrowright \text{E} \leftarrow \\ j \end{array} = \sum_j h_{ij'}^{\text{ext}}, \quad \begin{array}{l} j' \\ \curvearrowright \text{E} \rightarrow \\ i \end{array} = - \sum_i h_{ij'}^{\text{ext}}, \end{array} \quad (\text{C11})$$

$$\begin{array}{c} \begin{array}{l} j'_1 \quad i'_1 \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ j'_n \quad i'_n \end{array} = \langle \hat{a}_{i'_1} \dots \hat{a}_{i'_n} \hat{a}_{j'_n}^\dagger \dots \hat{a}_{j'_1}^\dagger \rangle_t^c, \quad \begin{array}{l} j' \quad i' \\ \bullet \rightarrow \bullet \end{array} = \langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t^c, \quad \begin{array}{l} j' \\ \bullet \rightarrow \\ \bullet \end{array}, \quad \begin{array}{l} i' \\ \bullet \rightarrow \\ \bullet \end{array}, \end{array} \quad (\text{C12})$$

where the last two graphs—*external vertices*—are involved in different contractions. In this notation Eq. (C10) is presented in Fig. 11. The result of applying operator $[\hat{D} + \hat{E}]$ is obvious. Here and further in the brackets one can see the number of terms which correspond to a presented unlabeled diagram. For \mathfrak{P}_k^y from (18) one derives the following:

$$\frac{d}{dt} \langle \hat{a}_{(c,k)} \hat{a}_{(v,k)}^\dagger \rangle_t^c + i[\varepsilon_{(c,k)} - \varepsilon_{(v,k)}] \langle \hat{a}_{(c,k)} \hat{a}_{(v,k)}^\dagger \rangle_t^c + i \left[\sum_j h_{(c,k)j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{(v,k)}^\dagger \rangle_t^c - \sum_i h_{i(v,k)}^{\text{ext}} \langle \hat{a}_{(c,k)} \hat{a}_i^\dagger \rangle_t^c \right] = 0, \quad (\text{C13})$$

$$\begin{aligned} i \sum_j h_{(c,k)j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{(v,k)}^\dagger \rangle_t^c &= i \left(-\frac{\mathcal{E}(t) d_{cv}}{\hbar} \right) \sum_{k'} \sum_j [\delta_{(c,k), (c,k')} \delta_{j, (v,k')} + \delta_{j, (c,k')} \delta_{(c,k), (v,k')}] \langle \hat{a}_j \hat{a}_{(v,k)}^\dagger \rangle_t^c \\ &= i \left(-\frac{\mathcal{E}(t) d_{cv}}{\hbar} \right) [1 - n_{v,k}^t], \end{aligned} \quad (\text{C14})$$

$$i \sum_i h_{i(v,k)}^{\text{ext}} \langle \hat{a}_{(c,k)} \hat{a}_i^\dagger \rangle_t^c = i \left(-\frac{\mathcal{E}(t) d_{cv}}{\hbar} \right) [1 - n_{c,k}^t]. \quad (\text{C15})$$

Taking into account the relation (11), in terms of polarization components we obtain

$$\hbar \frac{d}{dt} \tilde{\mathfrak{P}}_k^t + i \hbar [\varepsilon_{(c,k)} - \varepsilon_{(v,k)}] \tilde{\mathfrak{P}}_k^t + i \mathcal{E}(t) d_{cv} [n_{c,k}^t - n_{v,k}^t] = 0. \quad (\text{C16})$$

Thus, we derive the dynamical equation on the polarization component in the noninteracting case. The corresponding algebraic equation for the susceptibility component (32) is as follows:

$$\chi_I(\mathbf{k}, \omega) = \frac{d_{cv} [f_{(c,k)} - f_{(v,k)}]}{\hbar[\omega + i\delta - (\varepsilon_{(c,k)} - \varepsilon_{(v,k)})]} = -\frac{d_{cv} [1 - f_{(e,k)} - f_{(h,k)}]}{\hbar[\omega + i\delta - (\varepsilon_{(e,k)} + \varepsilon_{(h,k)})]}. \quad (\text{C17})$$

Also, instead of n we consider their equilibrium values—Fermi-Dirac distributions of carriers in valence and conductivity bands. The one-particle energies are defined as follows:

$$\hbar \varepsilon_{(e,k)} \equiv \hbar \varepsilon_{(c,k)} = E_g + \frac{\hbar^2 \mathbf{k}^2}{2m_c}, \quad \hbar \varepsilon_{(v,k)} = \frac{\hbar^2 \mathbf{k}^2}{2m_v}, \quad \hbar \varepsilon_{(h,k)} = \frac{\hbar^2 \mathbf{k}^2}{2m_h}. \quad (\text{C18})$$

2. One-particle dynamics

In this part we aim to derive the function $V_{1,1}[\langle 2 \rangle_{1\mathcal{E}}]$ from (25). For this purpose, however, we have to rewrite the commutator of $\hat{a}_{i'} \hat{a}_{j'}^\dagger$ and \hat{V} in terms of correlations. For the right-hand side of (12) we obtain

$$-i \langle [\hat{a}_{i'} \hat{a}_{j'}^\dagger, V] \rangle_t = -i \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} CM_1[i', j', i_1, i_2, j_2, j_1], \quad (\text{C19})$$

where for the convenience of the further calculations the two-operator-potential commutator was introduced:

$$CM_1[i', j', i_1, i_2, j_2, j_1] = \frac{1}{4} \left[\langle \hat{a}_{i'} \hat{a}_{j'}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t - \langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \hat{a}_{i'} \hat{a}_{j'}^\dagger \rangle_t \right]. \quad (\text{C20})$$

By means of the cluster expansion technique (14) the appeared difference of expectation values can be expanded as follows:

$$\begin{aligned}
 \langle \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t - \langle \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_j \hat{a}_i \hat{a}_i^\dagger \rangle_t &= \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c - \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t^c - \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_1} \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_2} \rangle_t^c \\
 &+ \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c - \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t^c + \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t^c \\
 &+ \langle \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_1} \rangle_t \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t^c - \langle \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_1} \rangle_t^c + \langle \hat{a}_i \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_1} \rangle_t \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t^c \\
 &+ \langle \hat{a}_i \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_1} \rangle_t \langle \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t^c - \langle \hat{a}_i \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c - \langle \hat{a}_i \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t^c \\
 &+ \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t^c + \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t^c - \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t^c \\
 &- \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t^c. \tag{C21}
 \end{aligned}$$

Due to the symmetry properties of potential (5) and correlations (17), the expression (C21) can be reduced to the combination of 3 unique terms (grouped by means of square brackets):

$$\begin{aligned}
 \langle \hat{a}_i \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t - \langle \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \hat{a}_i \hat{a}_i^\dagger \rangle_t \\
 = 4 \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \left[- \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c + \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t^c \right] \\
 + 2 \langle \hat{a}_i \hat{a}_{j_2} \hat{a}_i^\dagger \hat{a}_i^\dagger \rangle_t \left[\langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c + \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t^c \right] \\
 - 2 \langle \hat{a}_{j_1} \hat{a}_{j_2} \hat{a}_i^\dagger \hat{a}_i^\dagger \rangle_t \left[\langle \hat{a}_i \hat{a}_i^\dagger \rangle_t^c + \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t^c \right]. \tag{C22}
 \end{aligned}$$

This grouping will be clear later. Thus, taking into account (4) and (15) for commutator (C19) we find

$$\begin{aligned}
 -i \langle [\hat{a}_i \hat{a}_j^\dagger, V] \rangle_t &= -i \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[\frac{1}{2} \langle \hat{a}_i \hat{a}_{j_2} \hat{a}_i^\dagger \hat{a}_i^\dagger \rangle_t \delta_{j' j_1} \right. \\
 &- \frac{1}{2} \langle \hat{a}_{j_1} \hat{a}_{j_2} \hat{a}_i^\dagger \hat{a}_i^\dagger \rangle_t \delta_{i' i_1} \\
 &+ \langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \left[- \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c \right. \\
 &\left. \left. + \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t^c \right] \right]. \tag{C23}
 \end{aligned}$$

In terms of the schematic equation (23) we obtain the following parts:

$$\begin{aligned}
 V_{1,1}[\langle 2 \rangle_{(1)'}] \\
 = -i \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[\langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t \left[- \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c \right. \right. \\
 \left. \left. + \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t^c \right] \right], \tag{C24}
 \end{aligned}$$

$$\begin{aligned}
 V_{2,1}[\langle 2 \rangle_t^c] &= -i \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[\frac{1}{2} \langle \hat{a}_i \hat{a}_{j_2} \hat{a}_i^\dagger \hat{a}_i^\dagger \rangle_t \delta_{j' j_1} \right. \\
 &\left. - \frac{1}{2} \langle \hat{a}_{j_1} \hat{a}_{j_2} \hat{a}_i^\dagger \hat{a}_i^\dagger \rangle_t \delta_{i' i_1} \right]. \tag{C25}
 \end{aligned}$$

The expression (C24) allows us to write the equation on polarization dynamics within a one-particle approximation. Hence,

for (25) we find

$$\begin{aligned}
 \frac{d}{dt} \langle \hat{a}_i \hat{a}_j^\dagger \rangle_t^c + i[\varepsilon_{i'} - \varepsilon_{j'}] \langle \hat{a}_i \hat{a}_j^\dagger \rangle_t^c \\
 + i \left[\sum_j h_{ij}^{\text{ext}} \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t^c - \sum_i h_{ij}^{\text{ext}} \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t^c \right] \\
 = -i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[\langle \hat{a}_i^\dagger \hat{a}_{j_2} \rangle_t^c \left[- \langle \hat{a}_i \hat{a}_i^\dagger \rangle_t^c \langle \hat{a}_j^\dagger \hat{a}_{j_1} \rangle_t^c \right. \right. \\
 \left. \left. + \langle \hat{a}_i^\dagger \hat{a}_i \rangle_t^c \langle \hat{a}_j \hat{a}_j^\dagger \rangle_t^c \right] \right], \tag{C26}
 \end{aligned}$$

where the corresponding diagrammatic counterpart is depicted in Fig. 12. Let us obtain now the algebraic equation for susceptibility. For $\mathfrak{P}(\mathbf{k}, t)$ from (18) one has to treat $V_{1,1}[\langle 2 \rangle_{(1)'}^c]$:

$$\begin{aligned}
 V_{1,1}[\langle 2 \rangle_{(1)'}^c] \\
 = -i \sum_{q \neq 0} V_q \left[(n_{v, k+q}^t - n_{c, k+q}^t) \mathfrak{P}_k^t + (n_{c, k}^t - n_{v, k}^t) \mathfrak{P}_{k+q}^t \right] \\
 = i \left[\mathfrak{P}_k^t \sum_{q \neq k} V_{k-q} (n_{c, q}^t - n_{v, q}^t) + (n_{v, k}^t - n_{c, k}^t) \sum_{q \neq k} V_{k-q} \mathfrak{P}_q^t \right]. \tag{C27}
 \end{aligned}$$

Combining the previously obtained results and keeping in mind that $\mathfrak{P}_k^t = -\mathfrak{P}_k^t$, we obtain the well-known semiconductor Bloch equation [38]:

$$\begin{aligned}
 i \hbar \frac{d}{dt} \tilde{\mathfrak{P}}_k^t - [\hbar \epsilon_{(c, k)} - \hbar \epsilon_{(v, k)}] \tilde{\mathfrak{P}}_k^t \\
 = [n_{c, k}^t - n_{v, k}^t] \left[\mathcal{E}(t) d_{cv} + \sum_{q \neq k} V_{k-q} \tilde{\mathfrak{P}}_q^t \right], \tag{C28}
 \end{aligned}$$

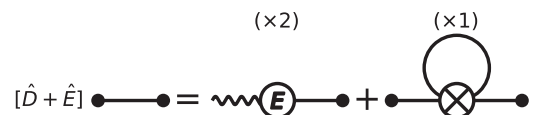


FIG. 12. The differential equation on one-particle correlations within one-particle approximation ($N = II$).

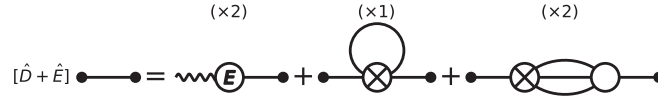


FIG. 13. The exact differential equation on one-particle correlation dynamics.

where the renormalized energies were introduced:

$$\hbar\epsilon_{(c,k)} = \hbar\epsilon_{(c,k)} - \sum_{q \neq k} V_{k-q} n_{c,q}^t, \quad (\text{C29})$$

$$\hbar\epsilon_{(v,k)} = \hbar\epsilon_{(v,k)} - \sum_{q \neq k} V_{k-q} n_{v,q}^t, \quad (\text{C30})$$

$$\hbar\epsilon_{(h,k)} = \hbar\epsilon_{(h,k)} - \sum_{q \neq k} V_{k-q} n_{h,q}^t. \quad (\text{C31})$$

The corresponding algebraic equation for susceptibility component $\chi(\mathbf{k}, \omega)$ reads

$$\begin{aligned} & \hbar[\omega + i\delta - [\epsilon_{(c,k)} - \epsilon_{(v,k)}]] \chi_{II}(\mathbf{k}, \omega) \\ &= [f_{(c,k)} - f_{(v,k)}] \left[d_{cv} + \sum_{q \neq k} V_{k-q} \chi_{II}(\mathbf{q}, \omega) \right]. \end{aligned} \quad (\text{C32})$$

This equation can be rewritten:

$$\chi_{II}(\mathbf{k}, \omega) = \Gamma_{II}(\mathbf{k}) \chi_I^R(\mathbf{k}, \omega), \quad (\text{C33})$$

where in terms of electrons and holes we have

$$\begin{aligned} \chi_I^R(\mathbf{k}, \omega) &= -\frac{d_{cv}[1 - f_{(e,k)} - f_{(h,k)}]}{\hbar[\omega + i\delta - (\epsilon_{(e,k)} + \epsilon_{(h,k)})]}, \\ \Gamma_{II}(\mathbf{k}) &= 1 + \frac{1}{d_{cv}} \sum_{q \neq k} \chi_I^R(\mathbf{q}, \omega) V_{k-q} \Gamma_{II}(\mathbf{q}). \end{aligned} \quad (\text{C34})$$

The introducing of the function Γ allows one to solve the integral equation (C33) by means of the matrix inversion approach.

3. Two-particle dynamics

The first line of the system (26) can be simply derived by means of the expression (C25). Adding this term to (C26), we obtain exact equation for one-particle correlations dynamics:

$$\begin{aligned} & \frac{d}{dt} \langle \hat{a}_i^\dagger \hat{a}_j^\dagger \rangle_t^c + i[\epsilon_{i'} - \epsilon_{j'}] \langle \hat{a}_i^\dagger \hat{a}_j^\dagger \rangle_t^c + i \left[\sum_j h_{i'j}^{\text{ext}} \langle \hat{a}_j \hat{a}_i^\dagger \rangle_t^c - \sum_i h_{ij'}^{\text{ext}} \langle \hat{a}_i^\dagger \hat{a}_j^\dagger \rangle_t^c \right] \\ &= -i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2} \rangle_t^c \left[\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{i_1}^\dagger \rangle_t^c - \langle \hat{a}_{i_1}^\dagger \hat{a}_{i_1} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_1} \rangle_t^c \right] + \frac{1}{2} \left[\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2} \hat{a}_{i_2}^\dagger \hat{a}_{j_1} \rangle_t^c \delta_{j'j_1} - \langle \hat{a}_{j_1} \hat{a}_{j_2} \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c \delta_{i'i_1} \right] \right], \end{aligned} \quad (\text{C35})$$

or in terms of the previously introduced quantities:

$$\left[\frac{d}{dt} + i[\epsilon_{i'} - \epsilon_{j'}] \right] \langle \hat{a}_i^\dagger \hat{a}_j^\dagger \rangle_t^c = -i \left[\sum_j T_{c, \{i'j\}, \{j,j'\}}^2 - \sum_i T_{c, \{ij'\}, \{i',i\}}^2 \right] - i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} CM_1[i', j', i_1, i_2, j_2, j_1], \quad (\text{C36})$$

where the corresponding graphical equation is presented in Fig. 13. Taking into account the potential form (B1), for polarization component from (C35) the dynamical equation looks as follows:

$$\begin{aligned} & \hbar \frac{d}{dt} \mathfrak{P}_k^i + i\hbar[\epsilon_{(c,k)} - \epsilon_{(v,k)}] \mathfrak{P}_k^i - i[n_{c,k}^t - n_{v,k}^t] \left[\mathcal{E}(t) d_{cv} - \sum_{q \neq k} V_{k-q} \mathfrak{P}_q^i \right] \\ &= i \sum_{k_2, q \neq 0} V_q \left[\langle \hat{a}_{(c,k-q)} \hat{a}_{(c,k_2)} \hat{a}_{(c,k_2-q)}^\dagger \hat{a}_{(v,k)}^\dagger \rangle_t^c + \langle \hat{a}_{(c,k-q)} \hat{a}_{(v,k_2)} \hat{a}_{(v,k_2-q)}^\dagger \hat{a}_{(v,k)}^\dagger \rangle_t^c \right. \\ & \quad \left. - \langle \hat{a}_{(c,k)} \hat{a}_{(v,k_2)} \hat{a}_{(v,k_2-q)}^\dagger \hat{a}_{(v,k+q)}^\dagger \rangle_t^c - \langle \hat{a}_{(c,k)} \hat{a}_{(c,k_2)} \hat{a}_{(c,k_2-q)}^\dagger \hat{a}_{(v,k+q)}^\dagger \rangle_t^c \right], \end{aligned} \quad (\text{C37})$$

while the corresponding algebraic equation for susceptibility component reads as

$$\begin{aligned} \chi(\mathbf{k}, \omega) &= \chi_I^R(\mathbf{k}, \omega) \left[1 + \frac{1}{d_{cv}} \sum_{q \neq k} V_{k-q} \chi(\mathbf{q}, \omega) \right] + \frac{1}{\mathcal{E}(\omega) \hbar[\omega + i\delta - (\epsilon_{(e,k)} + \epsilon_{(h,k)})]} \\ & \quad \times \sum_{k_2, q \neq 0} V_q \left[\langle \hat{a}_{(c,k-q)} \hat{a}_{(c,k_2)} \hat{a}_{(c,k_2-q)}^\dagger \hat{a}_{(v,k)}^\dagger \rangle_\omega^c + \langle \hat{a}_{(c,k-q)} \hat{a}_{(v,k_2)} \hat{a}_{(v,k_2-q)}^\dagger \hat{a}_{(v,k)}^\dagger \rangle_\omega^c \right. \\ & \quad \left. - \langle \hat{a}_{(c,k)} \hat{a}_{(v,k_2)} \hat{a}_{(v,k_2-q)}^\dagger \hat{a}_{(v,k+q)}^\dagger \rangle_\omega^c - \langle \hat{a}_{(c,k)} \hat{a}_{(c,k_2)} \hat{a}_{(c,k_2-q)}^\dagger \hat{a}_{(v,k+q)}^\dagger \rangle_\omega^c \right]. \end{aligned} \quad (\text{C38})$$

All the multiparticle effects are encoded in the second and third lines of (C38). Unfortunately, the exact form of two-particle correlations $\langle \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_i^\dagger \hat{a}_j^\dagger \rangle_\omega^c$ is unknown. However, by means of the Heisenberg equation of motion (12) and cluster expansion

technique (14) we can derive the exact dynamical equation for two-particle correlations, which schematically looks like

$$\hbar \frac{d}{dt} \langle 2 \rangle_t^c = T_2[\langle 2 \rangle_t^c] + V_{1,2}[\langle 3 \rangle_{(2)_t^c, (1)_t^c}] + V_{2,2}[\langle 3 \rangle_t^c]. \quad (\text{C39})$$

Having obtained Eq. (C39), we can simply obtain the result for the currently considered (26) and further approximations by neglecting the corresponding contributions from (C39).

There is no the equation of motion for the correlations themselves. However, one can apply the Heisenberg EOM (12) to four-operator expectation value and after that by means of the cluster expansion (14) derives the similar equation for two-particle correlations. From (16) we have

$$\langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c = \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t - \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c + \langle \hat{a}_{i_1} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \rangle_t^c. \quad (\text{C40})$$

It is reasonable to consider the following expression:

$$\begin{aligned} & \left[\frac{d}{dt} + i[\varepsilon_{i_1} + \varepsilon_{i_2} - \varepsilon_{j_2} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \\ &= \left[\frac{d}{dt} + i[\varepsilon_{i_1} + \varepsilon_{i_2} - \varepsilon_{j_2} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t \\ & - \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_2} - \varepsilon_{j_2}] \right] \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c - \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_1} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c \\ & + \langle \hat{a}_{i_1} \hat{a}_{j_2}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_2} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \rangle_t^c + \langle \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_1} - \varepsilon_{j_2}] \right] \langle \hat{a}_{i_1} \hat{a}_{j_2}^\dagger \rangle_t^c. \end{aligned} \quad (\text{C41})$$

In (C41) we know everything about the second and third lines [instead of derivatives for one-particle correlations we can use (C36)]. Thus, we have to handle the following part:

$$\left[\frac{d}{dt} + i[\varepsilon_{i_1} + \varepsilon_{i_2} - \varepsilon_{j_2} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t. \quad (\text{C42})$$

To understand what we have to obtain as output let us look back at Heisenberg equation:

$$\hbar \frac{d}{dt} \langle A \rangle_t + i \langle [A, H_{el,k}] \rangle_t = -i \langle [A, H_I] \rangle_t - i \langle [A, V] \rangle_t. \quad (\text{C43})$$

In fact, the left-hand side of (C43) coincides with (C42) up to \hbar . Therefore, we have to calculate now two expectation values of the commutators $[A, H_I]$ and $[A, V]$. The first one is trivial (we already found it for the considered type of quantity A):

$$-i \langle [\hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger, H_I] \rangle_t = -i \hbar \left[\sum_j (T_{\{i_1 j\}, \{j_2^2, j_2^2 j_1\}}^4 + T_{\{i_2 j\}, \{i_1 j, j_2^2 j_1\}}^4) - \sum_i (T_{\{i j_2\}, \{i_1 i_2, i j_1\}}^4 + T_{\{i j_1\}, \{i_1 i_2, j_2^2 i\}}^4) \right], \quad (\text{C44})$$

where T^4 can be expressed via correlations as follows:

$$T_{\{i_1 j\}, \{j_2^2, j_2^2 j_1\}}^4 = h_{i_1 j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t = h_{i_1 j}^{\text{ext}} [\langle \hat{a}_j \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c + \langle \hat{a}_j \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c - \langle \hat{a}_j \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \rangle_t^c], \quad (\text{C45})$$

while to calculate the second average within the used formalism we have to address to cluster expansion. For the two-particle case this gives:

$$\begin{aligned} -i \langle [A, V] \rangle_t &= -i \langle [\hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger, V] \rangle_t \\ &= -i \frac{1}{4} \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} [\langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t - \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2} \hat{a}_{j_1} \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t] \\ &= -i \frac{1}{4} \hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} [4 \langle \hat{a}_{i_1} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_2} \rangle_t^c + 4 \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_2} \rangle_t^c \\ & + 4 \langle \hat{a}_{i_1} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c - 4 \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{j_2} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c - 4 \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_2} \rangle_t^c \\ & + 4 \langle \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c - 4 \langle \hat{a}_{j_1} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c - 4 \langle \hat{a}_{i_1} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_2} \rangle_t^c \\ & - 4 \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2} \rangle_t^c + 4 \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{j_2} \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_2} \rangle_t^c \langle \hat{a}_{j_2} \hat{a}_{j_1} \rangle_t^c - 4 \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{j_1} \hat{a}_{j_2} \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{i_2} \rangle_t^c] \end{aligned}$$

$$\begin{aligned}
& + 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c \langle \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c - 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c + 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c \langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \\
& + 2\langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c - 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c \langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c - 4\langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \\
& + 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c - 4\langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c + 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \\
& + 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c \langle \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c + 2\langle \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \rangle_t^c + 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_1, i_1} \\
& - 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_1, i_2} + 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_1, i_2} - 4\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{i_1, i_2} \\
& - 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_2, i_2} - 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{i_2, i_2} - 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{j_1, j_1} \\
& + 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{j_1, j_2} - 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{j_1, j_2} + 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{j_1, j_2} \\
& + 4\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{j_2, j_2} - 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{j_2, j_2} - 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_1, i_2} \\
& - 2\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_2, i_2} - 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{j_1, j_2} - 2\langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1}^\dagger \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{j_2, j_2}] \\
& = -i\hbar \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} CM_2[i'_1, i'_2, j'_2, j'_1, i_1, i_2, j_2, j_1]. \tag{C46}
\end{aligned}$$

Thus, we know all the necessary expressions in terms of correlations for equation (C41). Let us present for completeness of the study, despite the cumbersomeness of all expressions, all terms entering (C41) via correlations:

$$\begin{aligned}
-\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_2} - \varepsilon_{j_2}] \right] \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c &= +i \left[\sum_j \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c T_{(i_2 j), (j j_2)}^2 - \sum_i \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c T_{(i j_2), (i_2 i)}^2 \right] \\
&+ i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c CM_1[i'_2, j'_2, i_1, i_2, j_2, j_1], \tag{C47}
\end{aligned}$$

$$\begin{aligned}
-\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_1} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c &= +i \left[\sum_j \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c T_{(i_1 j), (j j_1)}^2 - \sum_i \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c T_{(i j_1), (i_1 i)}^2 \right] \\
&+ i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c CM_1[i'_1, j'_1, i_1, i_2, j_2, j_1], \tag{C48}
\end{aligned}$$

$$\begin{aligned}
\langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_2} - \varepsilon_{j_1}] \right] \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c &= -i \left[\sum_j \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c T_{(i_2 j), (j j_1)}^2 - \sum_i \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c T_{(i j_1), (i_2 i)}^2 \right] \\
&- i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c CM_1[i'_2, j'_1, i_1, i_2, j_2, j_1], \tag{C49}
\end{aligned}$$

$$\begin{aligned}
\langle \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c \left[\frac{d}{dt} + i[\varepsilon_{i_1} - \varepsilon_{j_2}] \right] \langle \hat{a}_{i_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c &= -i \left[\sum_j \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c T_{(i_1 j), (j j_2)}^2 - \sum_i \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c T_{(i j_2), (i_1 i)}^2 \right] \\
&- i \sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \langle \hat{a}_{i_2}^\dagger \hat{a}_{j_1}^\dagger \rangle_t^c CM_1[i'_1, j'_2, i_1, i_2, j_2, j_1], \tag{C50}
\end{aligned}$$

and the last one is

$$\begin{aligned}
 \left[\frac{d}{dt} + i[\varepsilon_{i_1'} + \varepsilon_{i_2'} - \varepsilon_{j_2'} - \varepsilon_{j_1'}] \right] \langle \hat{a}_{i_1'} \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \hat{a}_{j_1'}^\dagger \rangle_t &= \frac{1}{\hbar} [-i\langle [A, H_I] \rangle_t - i\langle [A, V] \rangle_t] \\
 &= -i \left[\sum_j (T_{(i_1'j), (j'j_2'j_1')}^4 + T_{(i_2'j), (i_1'j_2'j_1')}^4) - \sum_i (T_{(i'j_2'), (i_1'i_2'j_1')}^4 + T_{(i'j_1'), (i_1'i_2'j_2i)}^4) \right] \\
 &\quad - i \left[\sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} CM_2[i_1', i_2', j_2', j_1', i_1, i_2, j_2, j_1] \right]. \tag{C51}
 \end{aligned}$$

Thus, for two-particle correlations one obtains

$$\begin{aligned}
 \left[\frac{d}{dt} + i[\varepsilon_{i_1'} + \varepsilon_{i_2'} - \varepsilon_{j_2'} - \varepsilon_{j_1'}] \right] \langle \hat{a}_{i_1'} \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \hat{a}_{j_1'}^\dagger \rangle_t^c &= -i \left[\sum_j [-\langle \hat{a}_{i_1'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(i_2'j), (j,j_2')}^2 - \langle \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i_1'j), (j,j_1')}^2 + \langle \hat{a}_{i_1'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i_2'j), (j,j_1')}^2 + \langle \hat{a}_{i_2'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(i_1'j), (j,j_2')}^2 \right. \\
 &\quad + T_{(i_1'j), (j'j_2'j_1')}^4 + T_{(i_2'j), (i_1'j_2'j_1')}^4] - \sum_i [-\langle \hat{a}_{i_1'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(ij_2'), (i_2',i)}^2 - \langle \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(ij_1'), (i_1',i)}^2 \\
 &\quad + \langle \hat{a}_{i_1'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i'j_1'), (i_2',i)}^2 + \langle \hat{a}_{i_2'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(ij_2'), (i_1',i)}^2 + T_{(ij_2'), (i_1'i_2',i'j_1')}^4 + T_{(i'j_1'), (i_1'i_2',j_2i)}^4] \\
 &\quad - i \left[\sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} [-\langle \hat{a}_{i_1'} \hat{a}_{j_1'}^\dagger \rangle_t^c CM_1[i_2', j_2', i_1, i_2, j_2, j_1] - \langle \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \rangle_t^c CM_1[i_1', j_1', i_1, i_2, j_2, j_1] \right. \\
 &\quad \left. + \langle \hat{a}_{i_1'} \hat{a}_{j_2'}^\dagger \rangle_t^c CM_1[i_2', j_1', i_1, i_2, j_2, j_1] + \langle \hat{a}_{i_2'} \hat{a}_{j_1'}^\dagger \rangle_t^c CM_1[i_1', j_2', i_1, i_2, j_2, j_1] + CM_2[i_1', i_2', j_2', j_1', i_1, i_2, j_2, j_1] \right]. \tag{C52}
 \end{aligned}$$

One can simplify each term in (C52):

$$\begin{aligned}
 \sum_j [-\langle \hat{a}_{i_1'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(i_2'j), (j,j_2')}^2 - \langle \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i_1'j), (j,j_1')}^2 + \langle \hat{a}_{i_1'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i_2'j), (j,j_1')}^2 + \langle \hat{a}_{i_2'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(i_1'j), (j,j_2')}^2 + T_{(i_1'j), (j'j_2'j_1')}^4 + T_{(i_2'j), (i_1'j_2'j_1')}^4] \\
 = \sum_j [h_{i_1'j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \hat{a}_{j_1'}^\dagger \rangle_t^c + h_{i_2'j}^{\text{ext}} \langle \hat{a}_{i_1'} \hat{a}_j \hat{a}_{j_2'}^\dagger \hat{a}_{j_1'}^\dagger \rangle_t^c] \\
 = \sum_j [T_{c, (i_1'j), (j'j_2'j_1')}^4 + T_{c, (i_2'j), (i_1'j_2'j_1')}^4], \tag{C53}
 \end{aligned}$$

where we introduce

$$T_{c, (i_1'j), (j'j_2'j_1')}^4 = h_{i_1'j}^{\text{ext}} \langle \hat{a}_j \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \hat{a}_{j_1'}^\dagger \rangle_t^c. \tag{C54}$$

This definition is being expanded to arbitrary correlations. Similarly for i summation one can find

$$\begin{aligned}
 \sum_i [-\langle \hat{a}_{i_1'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(ij_2'), (i_2',i)}^2 - \langle \hat{a}_{i_2'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i'j_1'), (i_1',i)}^2 + \langle \hat{a}_{i_1'} \hat{a}_{j_2'}^\dagger \rangle_t^c T_{(i'j_1'), (i_2',i)}^2 + \langle \hat{a}_{i_2'} \hat{a}_{j_1'}^\dagger \rangle_t^c T_{(ij_2'), (i_1',i)}^2 + T_{(ij_2'), (i_1'i_2',i'j_1')}^4 + T_{(i'j_1'), (i_1'i_2',j_2i)}^4] \\
 = \sum_i [T_{c, (i'j_1'), (i_1'i_2',j_2i)}^4 + T_{c, (ij_2'), (i_1'i_2',i'j_1')}^4]. \tag{C55}
 \end{aligned}$$

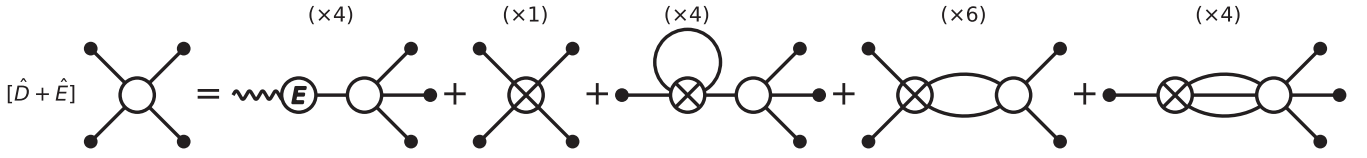


FIG. 14. The exact differential equation on two-particle correlation dynamics.

$$\begin{aligned}
 V_{2,2}[\langle 3 \rangle_t^c] = & -i\hbar \left[\sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[-\frac{1}{2} [\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_2} \hat{a}_{j_2} \hat{a}_{j_1}^\dagger \hat{a}_{j_1} \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_1, i_2}] + \frac{1}{2} [\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_1} \hat{a}_{j_2} \hat{a}_{j_1}^\dagger \hat{a}_{j_1} \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{i_2, i_2'}] \right. \right. \\
 & \left. \left. - \frac{1}{2} [\langle \hat{a}_{i_2}^\dagger \hat{a}_{i_1} \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_1} \hat{a}_{j_2}^\dagger \rangle_t^c \delta_{j_1, j_2}] + \frac{1}{2} [\langle \hat{a}_{i_2}^\dagger \hat{a}_{i_2} \hat{a}_{i_1} \hat{a}_{i_1} \hat{a}_{j_2} \hat{a}_{j_1}^\dagger \rangle_t^c \delta_{j_2, j_2'}] \right] \right]. \quad (\text{C61})
 \end{aligned}$$

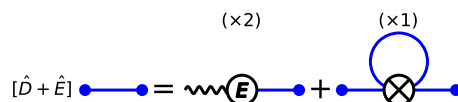
This exact equation on two-particle correlations dynamics has the following diagrammatic notation, which presented in Fig. 14. These results allow us to obtain the second line of the system (26):

$$\begin{aligned}
 & \left[\frac{d}{dt} + i[\varepsilon_{i_1} + \varepsilon_{i_2} - \varepsilon_{j_1} - \varepsilon_{j_2}] \right] \langle \hat{a}_{i_1} \hat{a}_{i_2} \hat{a}_{j_1}^\dagger \hat{a}_{j_2}^\dagger \rangle_t^c \\
 = & -i \left[\sum_{\substack{i_1, i_2 \\ j_1, j_2}} v_{i_1, i_2, j_1, j_2} \left[[\langle \hat{a}_{i_1}^\dagger \hat{a}_{i_1} \rangle_t^c \langle \hat{a}_{i_2}^\dagger \hat{a}_{i_2} \rangle_t^c \langle \hat{a}_{j_1}^\dagger \hat{a}_{j_1} \rangle_t^c \langle \hat{a}_{j_2}^\dagger \hat{a}_{j_2} \rangle_t^c - \langle \hat{a}_{j_1} \hat{a}_{j_1}^\dagger \rangle_t^c \langle \hat{a}_{j_2} \hat{a}_{j_2}^\dagger \rangle_t^c \langle \hat{a}_{i_1} \hat{a}_{i_1} \rangle_t^c \langle \hat{a}_{i_2} \hat{a}_{i_2} \rangle_t^c] \right] \right], \quad (\text{C62})
 \end{aligned}$$

which corresponds only to the first diagram of the right-hand side in Fig. 14. Thus, due to the possible combinations of zone indices $\{\lambda_1, \lambda_2, \lambda_3, \lambda_4\}$ we have 16 unique equations. The corresponding expressions for arbitrary momenta can be found in the Supplemental Material [51] as a Mathematica file. Here, we present the most relevant ones for the goals of the current approximation. The interesting combinations can be easily identified based on (C38). Taking into account all the adopted homogeneous approximations (31) and (30) we immediately see that such two-particle correlations on the basis of Eq. (C62) can be expressed in terms of \mathfrak{P}_k^t and $n_{\lambda, k}^t$ and consequently via of $\chi(\mathbf{k}, \omega)$. As usual, having obtained the differential equation we find the corresponding Fourier transform. For one of the terms entering (C38) from (C62) we derive

$$\begin{aligned}
 & \hbar \left[\frac{d}{dt} + i[\varepsilon_{(c, k-q)} + \varepsilon_{(c, k_2)} - \varepsilon_{(c, k_2-q)} - \varepsilon_{(v, k)}] \right] \langle \hat{a}_{(c, k-q)} \hat{a}_{(c, k_2)} \hat{a}_{(c, k_2-q)}^\dagger \hat{a}_{(v, k)}^\dagger \rangle_t^c \\
 = & i[\mathfrak{P}_k^t V_{k-k_2} [n_{c, k_2-q}^t [n_{c, k_2}^t + n_{c, k-q}^t - 1] - n_{c, k_2}^t n_{c, k-q}^t + \mathfrak{P}_{k_2-q}^{t, \dagger} \mathfrak{P}_{k-q}^t] \\
 & + \mathfrak{P}_{k_2}^{t, \dagger} V_{k-k_2} [n_{c, k-q}^t [1 - n_{c, k_2-q}^t - n_{v, k}^t] + n_{c, k_2-q}^t n_{v, k}^t - \mathfrak{P}_{k_2-q}^{t, \dagger} \mathfrak{P}_{k-q}^t] \\
 & + \mathfrak{P}_k^t V_{q'} [n_{c, k_2}^t n_{c, k-q}^t + n_{c, k_2-q}^t [1 - n_{c, k_2}^t - n_{c, k-q}^t] - \mathfrak{P}_{k_2-q}^{t, \dagger} \mathfrak{P}_{k_2}^t] \\
 & + \mathfrak{P}_{k-q}^t V_{q'} [n_{c, k_2}^t [-1 + n_{c, k_2-q}^t + n_{v, k}^t] - n_{c, k_2-q}^t n_{v, k}^t + \mathfrak{P}_{k_2-q}^{t, \dagger} \mathfrak{P}_{k_2}^t]]. \quad (\text{C63})
 \end{aligned}$$

All of these terms are responsible for scattering processes which generate two-particle correlations. According to our interest to determine the polarization dynamics within the linear in field approximation, we omit all the contributions which are quadratic


 FIG. 15. The diagrammatic representation of semiconductor-Bloch equation by means of which $\chi_{II}(\mathbf{k}, \omega)$ can be computed.

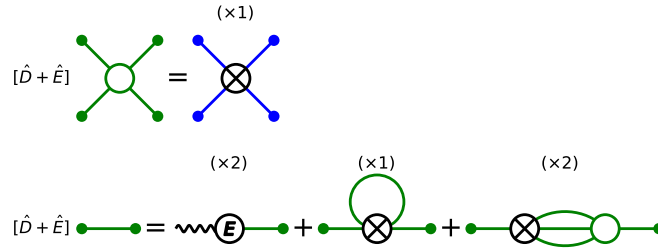


FIG. 16. The diagrammatic representation of equations on $\mathfrak{D}_{(\lambda_1, p_1), (\lambda_2, p_2), (\lambda_3, p_3), (\lambda_4, p_4)}^{\omega, III}$ and $\chi_{III}(\mathbf{k}, \omega)$, respectively.

or higher in the external field. Hence, the corresponding Fourier transform looks as

$$\begin{aligned} \langle \hat{a}_{(c, \mathbf{k}-\mathbf{q}')} \hat{a}_{(c, \mathbf{k}_2')} \hat{a}_{(c, \mathbf{k}_2'-\mathbf{q}')}^\dagger \hat{a}_{(v, \mathbf{k})}^\dagger \rangle_\omega^c &= \mathfrak{D}_{(c, \mathbf{k}-\mathbf{q}'), (c, \mathbf{k}_2'), (c, \mathbf{k}_2'-\mathbf{q}'), (v, \mathbf{k})}^{\omega, III} = F_{\mathfrak{D}, 1}(\chi_{II}, \mathbf{k} - \mathbf{q}', \mathbf{k}_2', \mathbf{k}_2' - \mathbf{q}', \mathbf{k}, \omega, \{cccv\}), \\ F_{\mathfrak{D}, 1}(\chi, \mathbf{k} - \mathbf{q}', \mathbf{k}_2', \mathbf{k}_2' - \mathbf{q}', \mathbf{k}, \omega, \{cccv\}) &= \frac{\mathcal{E}(\omega)}{\hbar[\omega + i\delta - [\varepsilon_{(c, \mathbf{k}-\mathbf{q}')} + \varepsilon_{(c, \mathbf{k}_2')} - \varepsilon_{(c, \mathbf{k}_2'-\mathbf{q}')} - \varepsilon_{(v, \mathbf{k})}]]} \\ &\times [\chi(\mathbf{k}, \omega) V_{\mathbf{k}-\mathbf{k}_2'} [-f_{(c, \mathbf{k}_2')} f_{(c, \mathbf{k}-\mathbf{q}')} + f_{(c, \mathbf{k}_2'-\mathbf{q}')} [-1 + f_{(c, \mathbf{k}_2')} + f_{(c, \mathbf{k}-\mathbf{q}')}]] \\ &+ \chi(\mathbf{k}_2', \omega) V_{\mathbf{k}-\mathbf{k}_2'} [f_{(c, \mathbf{k}-\mathbf{q}')} [1 - f_{(c, \mathbf{k}_2'-\mathbf{q}')} - f_{(v, \mathbf{k})}] + f_{(c, \mathbf{k}_2'-\mathbf{q}')} f_{(v, \mathbf{k})}] \\ &+ \chi(\mathbf{k}, \omega) V_{\mathbf{q}'} [f_{(c, \mathbf{k}_2')} f_{(c, \mathbf{k}-\mathbf{q}')} + f_{(c, \mathbf{k}_2'-\mathbf{q}')} [1 - f_{(c, \mathbf{k}_2')} - f_{(c, \mathbf{k}-\mathbf{q}')}]] \\ &+ \chi(\mathbf{k} - \mathbf{q}', \omega) V_{\mathbf{q}'} [f_{(c, \mathbf{k}_2')} [-1 + f_{(c, \mathbf{k}_2'-\mathbf{q}')} + f_{(v, \mathbf{k})}] - f_{(c, \mathbf{k}_2'-\mathbf{q}')} f_{(v, \mathbf{k})}], \end{aligned} \quad (C64)$$

where we took into account (32) and (11). For the second term with different zone indices combination we find

$$\begin{aligned} \langle \hat{a}_{(c, \mathbf{k}-\mathbf{q}')} \hat{a}_{(v, \mathbf{k}_2')} \hat{a}_{(v, \mathbf{k}_2'-\mathbf{q}')}^\dagger \hat{a}_{(v, \mathbf{k})}^\dagger \rangle_\omega^c &= \mathfrak{D}_{(c, \mathbf{k}-\mathbf{q}'), (v, \mathbf{k}_2'), (v, \mathbf{k}_2'-\mathbf{q}'), (v, \mathbf{k})}^{\omega, II} = F_{\mathfrak{D}, 1}(\chi_{II}, \mathbf{k} - \mathbf{q}', \mathbf{k}_2', \mathbf{k}_2' - \mathbf{q}', \mathbf{k}, \omega, \{cvvv\}), \\ F_{\mathfrak{D}, 1}(\chi, \mathbf{k} - \mathbf{q}', \mathbf{k}_2', \mathbf{k}_2' - \mathbf{q}', \mathbf{k}, \omega, \{cvvv\}) &= \frac{\mathcal{E}(\omega)}{\hbar[\omega + i\delta - [\varepsilon_{(c, \mathbf{k}-\mathbf{q}')} + \varepsilon_{(v, \mathbf{k}_2')} - \varepsilon_{(v, \mathbf{k}_2'-\mathbf{q}')} - \varepsilon_{(v, \mathbf{k})}]]} \\ &\times [\chi(\mathbf{k} - \mathbf{q}', \omega) V_{\mathbf{k}-\mathbf{k}_2'} [f_{(v, \mathbf{k}_2')} [1 - f_{(v, \mathbf{k})} - f_{(v, \mathbf{k}_2'-\mathbf{q}')}] + f_{(v, \mathbf{k})} f_{(v, \mathbf{k}_2'-\mathbf{q}')}]] \\ &+ \chi(\mathbf{k}_2' - \mathbf{q}', \omega) V_{\mathbf{k}-\mathbf{k}_2'} [f_{(v, \mathbf{k})} [-1 + f_{(c, \mathbf{k}-\mathbf{q}')} + f_{(v, \mathbf{k}_2')}] - f_{(c, \mathbf{k}-\mathbf{q}')} f_{(v, \mathbf{k}_2')}] \\ &+ \chi(\mathbf{k}, \omega) V_{\mathbf{q}'} [f_{(c, \mathbf{k}-\mathbf{q}')} f_{(v, \mathbf{k}_2')} + f_{(v, \mathbf{k}_2'-\mathbf{q}')} [1 - f_{(c, \mathbf{k}-\mathbf{q}')} - f_{(v, \mathbf{k}_2')}]] \\ &+ \chi(\mathbf{k} - \mathbf{q}', \omega) V_{\mathbf{q}'} [f_{(v, \mathbf{k}_2')} [-1 + f_{(v, \mathbf{k})} + f_{(v, \mathbf{k}_2'-\mathbf{q}')}] - f_{(v, \mathbf{k})} f_{(v, \mathbf{k}_2'-\mathbf{q}')}]. \end{aligned} \quad (C65)$$

The two remaining terms in (C38) can be derived easily by performing the momentum shift ($\mathbf{k} \rightarrow \mathbf{k} + \mathbf{q}$) in the already calculated expressions.

By the direct substitution of these expressions into (C38) we come to the system of multidimensional integral equations, obtaining an exact numerical solution for which is a huge challenge. In order to somehow tackle the problem, we can construct some iteration scheme. Let us describe it. Within the two-particle approximation dynamics each step of the procedure contains two operations. First, we find the answer for many-particle correlations ($\mathfrak{D}, \mathfrak{T}, \dots$); after that we construct the solution for the one-particle counterpart (χ). As a starting point for susceptibility we choose (C33) (see Fig. 15):

$$\chi_{II}(\mathbf{k}, \omega) = F_{\chi, 1}(\chi_{II}, \mathbf{k}, \omega),$$

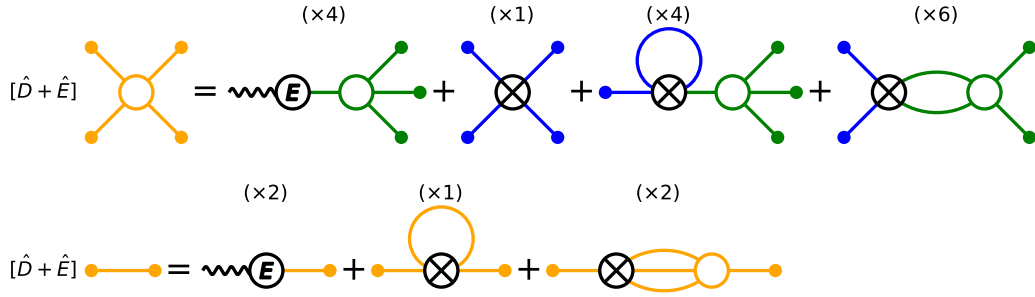
$$F_{\chi, 1}(\chi, \mathbf{k}, \omega) = \chi_I^R(\mathbf{k}, \omega) \left[1 + \frac{1}{d_{cv}} \sum_{\mathbf{q} \neq \mathbf{k}} V_{\mathbf{k}-\mathbf{q}} \chi(\mathbf{q}, \omega) \right], \quad (C66)$$

$$\mathfrak{D}^{\omega, 1} \equiv \mathfrak{D}^{\omega, 2} \equiv 0. \quad (C67)$$

As the next step one can find the susceptibility component with higher precision as follows (see Fig. 16):

$$\mathfrak{D}_{(\lambda_1, p_1), (\lambda_2, p_2), (\lambda_3, p_3), (\lambda_4, p_4)}^{\omega, III} = F_{\mathfrak{D}, 1}(\chi_{II}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}), \quad (C68)$$

$$\chi_{III}(\mathbf{k}, \omega) = F_{\chi, 1}(\chi_{III}, \mathbf{k}, \omega) + F_{\chi, 2}(\mathfrak{D}^{\omega, III}, \mathbf{k}, \omega), \quad (C69)$$


 FIG. 17. The diagrammatic representation of equations on Fourier transform of $\mathfrak{D}^{\omega,IV}$ and $\chi_{IV}(\mathbf{k}, \omega)$, respectively.

$$F_{\chi,2}(\mathfrak{D}^\omega, \mathbf{k}, \omega) = \frac{1}{\mathcal{E}(\omega)\hbar[\omega + i\delta - (\epsilon_{(c,\mathbf{k})} - \epsilon_{(v,\mathbf{k})})]} \sum_{\mathbf{k}'_2, \mathbf{q}' \neq 0} V_{\mathbf{q}'} [\mathfrak{D}_{(c,\mathbf{k}-\mathbf{q}'),(c,\mathbf{k}'_2),(c,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k})}^\omega + \mathfrak{D}_{(c,\mathbf{k}-\mathbf{q}'),(v,\mathbf{k}'_2),(v,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k})}^\omega - \mathfrak{D}_{(c,\mathbf{k}),(v,\mathbf{k}'_2),(v,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k}+\mathbf{q}')}^\omega - \mathfrak{D}_{(c,\mathbf{k}),(c,\mathbf{k}'_2),(c,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k}+\mathbf{q}')}^\omega]. \quad (\text{C70})$$

With these expressions the system (26) can be considered as a completely covered one.

As the next step, say to describe the system (28), we have to include the pure two-particle correlations into the right-hand side of their own dynamical equations. The formal equation for the susceptibility component in this case looks like

$$\chi_{IV}(\mathbf{k}, \omega) = F_{\chi,1}(\chi_{IV}, \mathbf{k}, \omega) + F_{\chi,2}(\mathfrak{D}^{\omega,IV}, \mathbf{k}, \omega). \quad (\text{C71})$$

Thus, in order to move forward, one has to define the form of the functions $\mathfrak{D}^{\omega,IV}$. The general structure of the corresponding exact function is highly transparent:

$$\mathfrak{D}_{(\lambda_1, \mathbf{p}_1), (\lambda_2, \mathbf{p}_2), (\lambda_3, \mathbf{p}_3), (\lambda_4, \mathbf{p}_4)}^\omega = F_{\mathfrak{D},1}(\chi, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}) + F_{\mathfrak{D},2}(\mathfrak{D}^\omega, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}) + F_{\mathfrak{D},3}(\mathfrak{T}^\omega, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}). \quad (\text{C72})$$

It is quite obvious that $F_{\mathfrak{D},2}$ stems from (C58) and (C60), while $F_{\mathfrak{D},3}$ is derived from (C61). It should be noted that without exceeding the two-particle approximation the computation of the term $F_{\mathfrak{D},3}$ has to be equated to zero. Following the suggested iterative scheme, for $\mathfrak{D}^{\omega,IV}$ we obtain (see Fig. 17)

$$\mathfrak{D}_{(\lambda_1, \mathbf{p}_1), (\lambda_2, \mathbf{p}_2), (\lambda_3, \mathbf{p}_3), (\lambda_4, \mathbf{p}_4)}^{\omega,IV} = F_{\mathfrak{D},1}(\chi_{II}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}) + F_{\mathfrak{D},2}(\mathfrak{D}^{\omega,III}, \mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \omega, \{\lambda_1 \lambda_2 \lambda_3 \lambda_4\}). \quad (\text{C73})$$

Thus, the last function which we have to derive is $F_{\mathfrak{D},2}$. These functions for arbitrary zone indices and momenta combinations are presented in the Supplemental Material [51] as a Mathematica file. To get an idea about the structure of this function, however, we demonstrate here only one particular case. Let us analyze first the contributions connected with the external field. For the particular zone indices from

$$\left[\frac{d}{dt} + i[\varepsilon_{i'_1} + \varepsilon_{i'_2} - \varepsilon_{j'_2} - \varepsilon_{j'_1}] \right] \langle \hat{a}_{i'_1} \hat{a}_{i'_2} \hat{a}_{j'_2}^\dagger \hat{a}_{j'_1}^\dagger \rangle_t^c = -i \left[\sum_j [T_{c, \{i'_1 j\}, \{j'_2 j'_1\}}^4 + T_{c, \{i'_2 j\}, \{i'_1 j'_2 j'_1\}}^4] - \sum_i [T_{c, \{i j'_1\}, \{i'_1 i'_2 j'_2 i\}}^4 + T_{c, \{i j'_2\}, \{i'_1 i'_2 i j'_1\}}^4] \right] \quad (\text{C74})$$

for combination $\{c, c, c, v\}$ we have

$$\left[\frac{d}{dt} + i[\varepsilon_{(c,\mathbf{k}-\mathbf{q})} + \varepsilon_{(c,\mathbf{k}_2)} - \varepsilon_{(c,\mathbf{k}_2-\mathbf{q})} - \varepsilon_{(v,\mathbf{k})}] \right] \langle \hat{a}_{(c,\mathbf{k}-\mathbf{q})} \hat{a}_{(c,\mathbf{k}_2)} \hat{a}_{(c,\mathbf{k}_2-\mathbf{q})}^\dagger \hat{a}_{(v,\mathbf{k})}^\dagger \rangle_t^c = \frac{i}{\hbar} d_{cv} \mathcal{E}(t) [-\langle \hat{a}_{(c,\mathbf{k}-\mathbf{q})} \hat{a}_{(c,\mathbf{k}_2)} \hat{a}_{(c,\mathbf{k}_2-\mathbf{q})}^\dagger \hat{a}_{(c,\mathbf{k})}^\dagger \rangle_t^c - \langle \hat{a}_{(c,\mathbf{k}-\mathbf{q})} \hat{a}_{(c,\mathbf{k}_2)} \hat{a}_{(v,\mathbf{k}_2-\mathbf{q})}^\dagger \hat{a}_{(v,\mathbf{k})}^\dagger \rangle_t^c + \langle \hat{a}_{(c,\mathbf{k}-\mathbf{q})} \hat{a}_{(v,\mathbf{k}_2)} \hat{a}_{(c,\mathbf{k}_2-\mathbf{q})}^\dagger \hat{a}_{(v,\mathbf{k})}^\dagger \rangle_t^c + \langle \hat{a}_{(v,\mathbf{k}-\mathbf{q})} \hat{a}_{(c,\mathbf{k}_2)} \hat{a}_{(c,\mathbf{k}_2-\mathbf{q})}^\dagger \hat{a}_{(v,\mathbf{k})}^\dagger \rangle_t^c]. \quad (\text{C75})$$

The structure of the other 15 equations is similar. All of them can be schematically written as follows:

$$\begin{aligned} & \{\{cccc\}, \{-\{cccv\}, -\{ccvc\}, +\{cvcc\}, +\{vccc\}\}\}, \quad \{\{cccv\}^*, \{-\{cccc\}, -\{ccvv\}, +\{cvvc\}, +\{vccv\}\}\}, \\ & \{\{ccvc\}, \{-\{ccvv\}, -\{cccc\}, +\{cvvc\}, +\{vccv\}\}\}, \quad \{\{cvcc\}, \{-\{cvvc\}, -\{cvvc\}, +\{cccc\}, +\{vccc\}\}\}, \\ & \{\{vccc\}, \{-\{vccv\}, -\{vccv\}, +\{vvcc\}, +\{cccc\}\}\}, \quad \{\{ccvv\}, \{-\{ccvc\}, -\{cccv\}, +\{cvvv\}, +\{vccv\}\}\}, \end{aligned}$$

$$\begin{aligned}
& \{\{cvvc\}, \{-\{cvvv\}, -\{cvcc\}, +\{cvcc\}, +\{vccc\}\}\}, \quad \{\{vvcc\}, \{-\{vvvc\}, -\{vvvc\}, +\{vccc\}, +\{cvcc\}\}\}, \\
& \{\{cvvv\}^*, \{-\{cvvc\}, -\{cvvc\}, +\{ccvv\}, +\{vvvv\}\}\}, \quad \{\{vvvc\}, \{-\{vvvv\}, -\{vvcc\}, +\{vcvc\}, +\{cvvc\}\}\}, \\
& \{\{vvvc\}, \{-\{vvcc\}, -\{vvvv\}, +\{vccv\}, +\{cvvc\}\}\}, \quad \{\{vcvv\}, \{-\{vcvc\}, -\{vccv\}, +\{vvvv\}, +\{ccvv\}\}\}, \\
& \{\{vcvc\}, \{-\{vcvv\}, -\{vccc\}, +\{vvvc\}, +\{ccvc\}\}\}, \quad \{\{cvvc\}, \{-\{cvcc\}, -\{cvvv\}, +\{cccv\}, +\{vvvc\}\}\}, \\
& \{\{vccv\}, \{-\{vccc\}, -\{vcvv\}, +\{vvcv\}, +\{cccv\}\}\}, \quad \{\{vvvv\}, \{-\{vvvc\}, -\{vvcv\}, +\{vcvv\}, +\{cvvv\}\}\}. \quad (C76)
\end{aligned}$$

We marked the combinations that are interesting for the purposes of the present work by a star sign. Also, some functions are in boldface type. It turns out that only these correlations possess the field-independent (via polarization) contributions when we include into consideration the terms connected with Coulomb interaction. For example, Eq. (C62) for $\{c, c, c, c\}$ reads as

$$\begin{aligned}
& \hbar \left[\frac{d}{dt} + i[\varepsilon_{(c,k-q')} + \varepsilon_{(c,k'_2)} - \varepsilon_{(c,k'_2-q')} - \varepsilon_{(c,k)}] \right] \langle \hat{a}_{(c,k-q')} \hat{a}_{(c,k'_2)} \hat{a}_{(c,k'_2-q')}^\dagger \hat{a}_{(c,k)}^\dagger \rangle_t^c \\
& = i[V_{k-k_2}[-n'_{c,k}[-1 + n'_{c,k-q'}]n'_{c,k_2-q'} + n'_{c,k_2}[-n'_{c,k}n'_{c,k_2-q'} + n'_{c,k-q'}[-1 + n'_{c,k} + n'_{c,k_2-q'}]]] \\
& \quad - V_{q'}[-n'_{c,k}[-1 + n'_{c,k-q'}]n'_{c,k_2-q'} + n'_{c,k_2}[-n'_{c,k}n'_{c,k_2-q'} + n'_{c,k-q'}[-1 + n'_{c,k} + n'_{c,k_2-q'}]]]. \quad (C77)
\end{aligned}$$

Keeping in mind the presence of $\mathcal{E}(t)$ as a multiplier in (C75) as well as the adopted limitation of analyzing only linear in field effects one can conclude that for such *boldface* combinations it is enough to consider only constant contributions with their subsequent substitution into the equations for $\{c, c, c, c\}$ and $\{c, v, v, v\}$. The deriving of the steady-state solution from (C77) is obvious:

$$\begin{aligned}
& \mathfrak{D}_{(c,k-q'),(c,k'_2),(c,k'_2-q'),(c,k)}^{III} \\
& = \langle \hat{a}_{(c,k-q')} \hat{a}_{(c,k'_2)} \hat{a}_{(c,k'_2-q')}^\dagger \hat{a}_{(c,k)}^\dagger \rangle^c = \frac{1}{\hbar[\varepsilon_{(c,k-q')} + \varepsilon_{(c,k'_2)} - \varepsilon_{(c,k'_2-q')} - \varepsilon_{(c,k)} + i\delta]} \\
& \quad \times [V_{k-k_2}[-f_{(c,k)}[-1 + f_{(c,k-q')}]f_{(c,k_2-q')} + f_{(c,k_2)}[-f_{(c,k)}f_{(c,k_2-q')} + f_{(c,k-q')}[-1 + f_{(c,k)} + f_{(c,k_2-q')}]]] \\
& \quad - V_{q'}[-f_{(c,k)}[-1 + f_{(c,k-q')}]f_{(c,k_2-q')} + f_{(c,k_2)}[-f_{(c,k)}f_{(c,k_2-q')} + f_{(c,k-q')}[-1 + f_{(c,k)} + f_{(c,k_2-q')}]]. \quad (C78)
\end{aligned}$$

Thus, combining Eqs. (C60) and (C74) and applying the Fourier transform within the linear in the external field approximation, we come to the following relation:

$$\begin{aligned}
& F_{\mathfrak{D},2}(\mathfrak{D}^\omega, \mathbf{k} - \mathbf{q}', \mathbf{k}'_2, \mathbf{k}'_2 - \mathbf{q}', \mathbf{k}, \omega, \{cvvv\}) \\
& = \frac{1}{\hbar[\omega + i\delta - [\varepsilon_{(c,k-q')} + \varepsilon_{(v,k'_2)} - \varepsilon_{(v,k'_2-q')} - \varepsilon_{(v,k)}]]} \\
& \quad \times [d_{cv}\mathcal{E}(\omega)[\mathfrak{D}_{(c,k-q'),(c,k'_2),(c,k'_2-q'),(c,k)} - \mathfrak{D}_{(c,k-q'),(v,k'_2),(c,k'_2-q'),(v,k)} - \mathfrak{D}_{(v,k-q'),(c,k'_2),(c,k'_2-q'),(v,k)}] \\
& \quad + \sum_{q''} V_{q''}[\mathfrak{D}_{(c,k-q'+q''),(c,k'_2),(c,k'_2-q''),(v,k+q'')}^\omega f_{(c,k-q')} - \mathfrak{D}_{(c,k-q'+q''),(c,k'_2-q''),(c,k'_2-q''),(v,k)}^\omega [-1 + f_{(c,k'_2)} + f_{(c,k-q')}] \\
& \quad + \mathfrak{D}_{(c,k'_2+q''),(c,k-q'),(c,k'_2-q'+q''),(v,k)}^\omega [-f_{(c,k'_2)} + f_{(c,k'_2-q')}] + \mathfrak{D}_{(c,k'_2),(c,k-q'+q''),(c,k'_2-q'+q''),(v,k)}^\omega [-f_{(c,k-q')} + f_{(c,k'_2-q')}] \\
& \quad - \mathfrak{D}_{(c,k-q'+q''),(c,k'_2),(c,k'_2-q''),(v,k+q'')}^\omega [f_{(v,k)}] + \mathfrak{D}_{(c,k'_2+q''),(c,k-q'),(c,k'_2-q''),(v,k+q'')}^\omega [-f_{(c,k'_2)} + f_{(v,k)}] \\
& \quad + \mathfrak{D}_{(c,k-q'),(c,k'_2),(c,k'_2-q'+q''),(v,k-q'')}^\omega [-1 + f_{(c,k'_2-q')} + f_{(v,k)}] \\
& \quad + [\mathfrak{D}_{(c,k-q'),(c,k'_2),(c,k-q''),(c,k'_2-q'+q'')}^\omega + \mathfrak{D}_{(c,k'_2+q''),(c,k-q'),(c,k+q''),(c,k'_2-q')}^\omega - \mathfrak{D}_{(c,k-q'+q''),(c,k'_2),(c,k+q''),(c,k'_2-q')}^\omega] \mathfrak{P}_k^\omega \\
& \quad - [\mathfrak{D}_{(c,k-q'),(v,k'_2+q''),(c,k'_2-q''),(v,k+q'')}^\omega + \mathfrak{D}_{(c,k-q'),(v,k'_2+q''),(c,k'_2-q'+q''),(v,k)}^\omega - \mathfrak{D}_{(c,k-q'+q''),(v,k'_2-q''),(c,k'_2-q''),(v,k)}^\omega] \mathfrak{P}_{k'_2}^\omega \\
& \quad + [\mathfrak{D}_{(c,k'_2),(v,k-q'+q''),(c,k'_2-q''),(v,k+q'')}^\omega + \mathfrak{D}_{(c,k'_2),(v,k-q'+q''),(c,k'_2-q'+q''),(v,k)}^\omega - \mathfrak{D}_{(c,k'_2+q''),(v,k-q'+q''),(c,k'_2-q''),(v,k)}^\omega] \mathfrak{P}_{k-q''}^\omega \\
& \quad - \mathfrak{D}_{(c,k-q'),(c,k'_2),(c,k'_2-q''),(c,k)}^\omega \mathfrak{P}_{k+q''}^\omega + \mathfrak{D}_{(c,k-q'),(v,k'_2),(c,k'_2-q''),(v,k)}^\omega \mathfrak{P}_{k'_2+q''}^\omega - \mathfrak{D}_{(c,k'_2),(v,k-q'),(c,k'_2-q''),(v,k)}^\omega \mathfrak{P}_{k-q'+q''}^\omega] \\
& \quad + V_{q'} \sum_{q''} [[-\mathfrak{D}_{(c,k-q'),(v,q''),(v,-q'+q''),(v,k)}^\omega + \mathfrak{D}_{(c,q''),(c,k-q'),(c,-q'+q''),(v,k)}^\omega][f_{(c,k'_2)} - f_{(c,k'_2-q')}] \\
& \quad + [\mathfrak{D}_{(c,k'_2),(v,q''),(c,k'_2-q''),(v,q'+q'')}^\omega - \mathfrak{D}_{(c,q''),(c,k'_2),(c,k'_2-q''),(c,q'+q'')}^\omega][\mathfrak{P}_k^\omega - \mathfrak{P}_{k-q''}^\omega] \\
& \quad + V_{k-k'_2} \sum_{q''} [[-\mathfrak{D}_{(c,q''),(c,k'_2),(c,-k+q''+k'_2),(v,k)}^\omega + \mathfrak{D}_{(c,k'_2),(v,q''),(v,-k+q''+k'_2),(v,k)}^\omega][f_{(c,k-q')} - f_{(c,k'_2-q')}] \\
& \quad - [\mathfrak{D}_{(c,k-q'),(v,q''),(c,k'_2-q''),(v,k-k'_2+q'')}^\omega + \mathfrak{D}_{(c,q''),(c,k-q'),(c,k-k'_2+q''),(c,k'_2-q'')}^\omega][\mathfrak{P}_k^\omega - \mathfrak{P}_{k'_2}^\omega]]. \quad (C79)
\end{aligned}$$

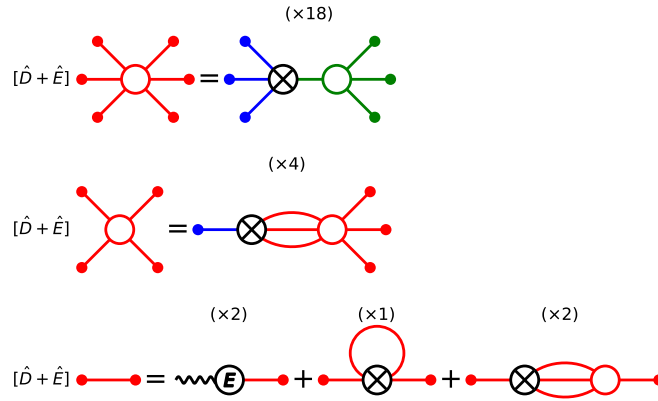


FIG. 19. The diagrammatic representation of equations on one-, two-, and three-particle correlations within the adopted approximation which needs to obtain χ_V behavior.

terms of diagrams, the last approximation which we consider in this article is shown in Fig. 19. Thus, we have to obtain the analytical expressions for the first diagram of the right-hand side of the first line in Fig. 19 and also for the last graph from the right-hand side of the second line when the particular set of momenta arguments is determined. In terms of the previously used functions this diagram containing the pure three-particle correlation bubble partly corresponds to $F_{\mathcal{D},3}$ from (C72) which in turn appeared from (C61). This function is used further in order to calculate $\mathcal{D}^{\omega,V}$. To get an idea about the expression structure we demonstrate here one of the four relevant combinations. This term reads as follows:

$$\begin{aligned}
 & F_{\mathcal{D},3}(\mathfrak{T}^{\omega}, \mathbf{k} - \mathbf{q}', \mathbf{k}'_2, \mathbf{k}'_2 - \mathbf{q}', \mathbf{k}, \omega, \{cccv\}) \\
 &= - \frac{1}{\hbar[\omega + i\delta - [\varepsilon_{(c,\mathbf{k}-\mathbf{q}')} + \varepsilon_{(v,\mathbf{k}'_2)} - \varepsilon_{(v,\mathbf{k}'_2-\mathbf{q}')} - \varepsilon_{(v,\mathbf{k})}]]} \\
 & \times \left[\sum_{\mathbf{q}'', \mathbf{k}''_2} V_{\mathbf{q}''} [\mathfrak{T}^{\omega}_{(c,\mathbf{k}''_2), (c,\mathbf{k}-\mathbf{q}'), (c,\mathbf{k}'_2), (c,\mathbf{k}'_2-\mathbf{q}'), (c,\mathbf{k}'_2+\mathbf{q}''), (v,\mathbf{k}-\mathbf{q}'')} + \mathfrak{T}^{\omega}_{(c,\mathbf{k}''_2), (c,\mathbf{k}-\mathbf{q}'), (c,\mathbf{k}'_2), (c,\mathbf{k}'_2-\mathbf{q}'-\mathbf{q}''), (c,\mathbf{k}'_2+\mathbf{q}''), (v,\mathbf{k})} \right. \\
 & - \mathfrak{T}^{\omega}_{(c,\mathbf{k}'_2+\mathbf{q}''), (c,\mathbf{k}-\mathbf{q}'), (c,\mathbf{k}'_2), (c,\mathbf{k}'_2+\mathbf{q}''), (c,\mathbf{k}'_2-\mathbf{q}'), (v,\mathbf{k})} + \mathfrak{T}^{\omega}_{(c,\mathbf{k}-\mathbf{q}'+\mathbf{q}''), (c,\mathbf{k}'_2), (c,\mathbf{k}'_2), (c,\mathbf{k}'_2-\mathbf{q}'), (v,\mathbf{k})} \\
 & + \mathfrak{T}^{\omega}_{(v,\mathbf{k}'_2), (c,\mathbf{k}'_2), (c,\mathbf{k}-\mathbf{q}'), (v,\mathbf{k}+\mathbf{q}''), (v,\mathbf{k}'_2-\mathbf{q}''), (c,\mathbf{k}'_2-\mathbf{q}')} - \mathfrak{T}^{\omega}_{(v,\mathbf{k}'_2), (c,\mathbf{k}'_2), (c,\mathbf{k}-\mathbf{q}'), (v,\mathbf{k}'_2+\mathbf{q}''), (v,\mathbf{k}), (c,\mathbf{k}'_2-\mathbf{q}')} \\
 & \left. + \mathfrak{T}^{\omega}_{(v,\mathbf{k}'_2), (c,\mathbf{k}'_2+\mathbf{q}''), (c,\mathbf{k}-\mathbf{q}'), (v,\mathbf{k}'_2+\mathbf{q}''), (v,\mathbf{k}), (c,\mathbf{k}'_2-\mathbf{q}')} - \mathfrak{T}^{\omega}_{(v,\mathbf{k}'_2), (c,\mathbf{k}-\mathbf{q}'+\mathbf{q}''), (c,\mathbf{k}'_2), (v,\mathbf{k}'_2+\mathbf{q}''), (v,\mathbf{k}), (c,\mathbf{k}'_2-\mathbf{q}')} \right]. \quad (\text{C81})
 \end{aligned}$$

Further we include into the equation on χ_V only the following difference:

$$F_{\mathcal{D},3}(\mathfrak{T}^{\omega}, \mathbf{k} - \mathbf{q}', \mathbf{k}'_2, \mathbf{k}'_2 - \mathbf{q}', \mathbf{k}, \omega, \{cccv\}) - F_{\mathcal{D},3}(\mathfrak{T}^{\omega}, \mathbf{k}, \mathbf{k}'_2, \mathbf{k}'_2 - \mathbf{q}', \mathbf{k} + \mathbf{q}', \omega, \{cccv\}), \quad (\text{C82})$$

believing that a such combination plays a dominant role when the density of electrons is much higher than its counterpart for holes. This expression allows us to understand for which combinations of zone indices we have to calculate functions \mathfrak{T} . The equation on three-particle correlations even within the considered approximation (only the 1d diagram) is dramatically lengthy. In such a situation we left only the terms which are no higher than $\sim \mathcal{D} \cdot f$, where f is the Fermi-Dirac distribution. We supply the files where these functions can be found.

APPENDIX D: NUMERICS

In this section we specify all the analytical results in the case of two-dimensional systems ($D = 2$).

1. Basic formulas and Bloch equation

First, we have to define the summation over the momentum space. It can be replaced by the integral evaluated in polar coordinates. Thus, in two dimensions for arbitrary function $g(\mathbf{k})$ we use

$$\sum_{\mathbf{k}} g(\mathbf{k}) = \left(\frac{L}{2\pi}\right)^2 \int_0^{2\pi} d\varphi \int_0^{\infty} g(\mathbf{k}) k dk, \quad (\text{D1})$$

where throughout the computations for simplicity we assume $L = 1$. Among the several quantities which have to be calculated, the first ones are chemical potentials of electrons and holes. For this purpose, one can address the following simple consideration:

$$n = \sum_{\mathbf{k}} f_{\mathbf{k}}. \quad (\text{D2})$$

Taking into account (D1) and keeping in mind that each summation contains an extra factor connected with spin structure, we come to the following answer which leads to the well-known expression

$$\mu_e = k_b T \ln(e^{\hbar^2 \beta \pi n_e / m_e} - 1) + E_g,$$

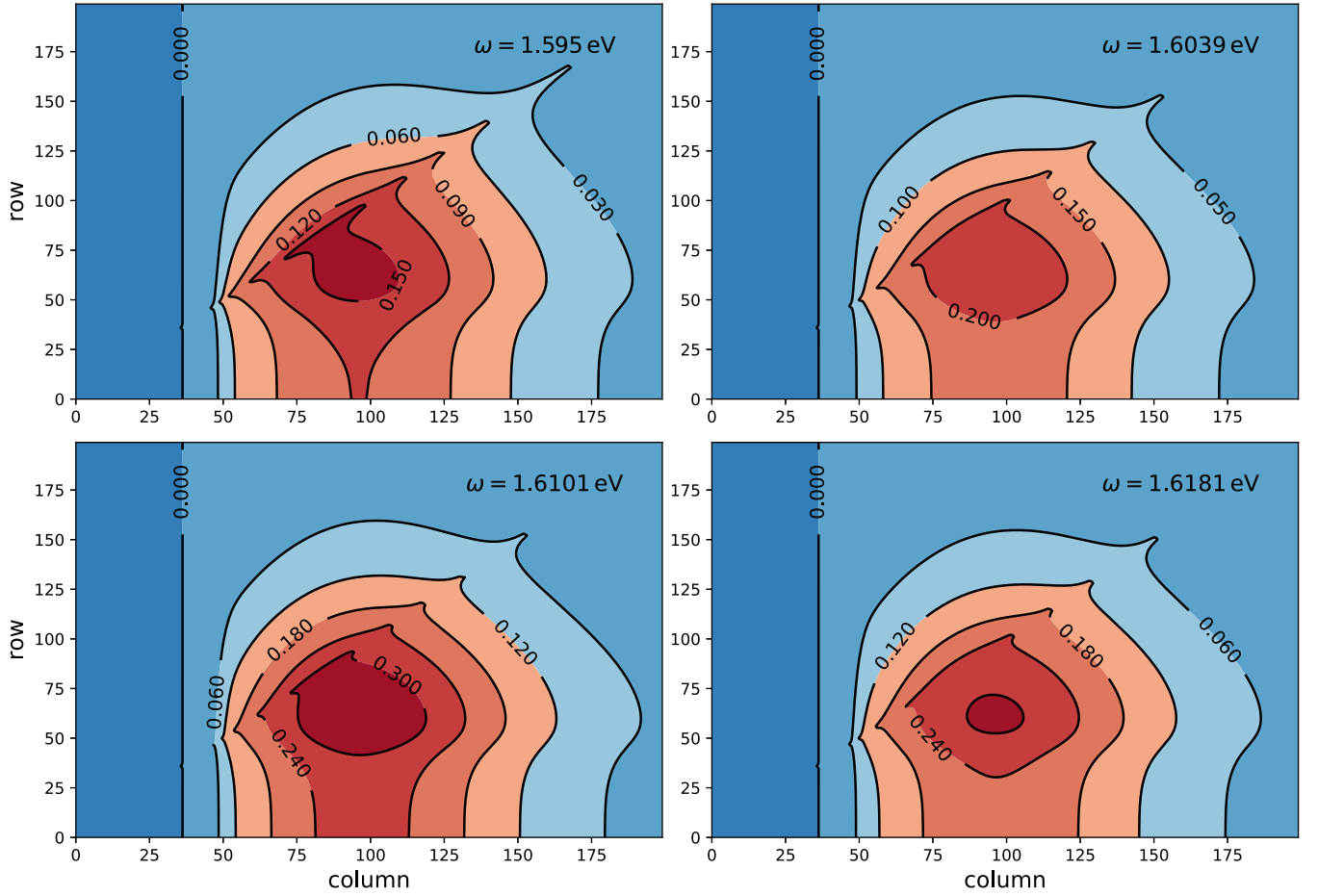


FIG. 20. The behavior of real part of matrix coefficients $\hat{M}_{i,j}^{-1} - \delta_{ij}$ for different values of frequency ω . This matrix was used to calculate the absorption in Fig. 10. The values of physical parameters including temperature and carrier densities can be found also there.

$$\mu_h = k_b T \ln(e^{\hbar^2 \beta \pi n_e / m_h} - 1). \quad (\text{D3})$$

This simple expression, of course, does not take into account the intrinsic features of a particular semiconductor and does not pretend to give a highly accurate numerical estimate for chemical potential. In order to tackle the internal structure of a sample one needs to address the more sophisticated relation. For example, in case of TMD MLs such a dependence was obtained recently (see, e.g., Appendix B in Ref. [56]). At this step the authors of the present paper decide not to complicate the consideration even more, believing that Eq. (D3) contains the main features of 2d semiconductors. It should be noted also that in practice we use some finite value k_{\max} as the upper limit in momentum integration at (D1). This number is extracted by means of the following procedure. First, we set some starting value, say k_{in} , where both carrier densities are x times smaller than at the origin ($k = 0$). Then, we calculate $\chi_{II}(k_{\text{in}}, \omega)$; if the obtained number is not equal to zero (within the machine precision) we increase the value of k_{in} by some reasonable factor and again analyze $\chi_{II}(k_{\text{in}}, \omega)$. We repeat these steps till the moment when we obtain $\chi_{II}(k_{\text{in}}, \omega) = 0$. The corresponding value of k_{in} we accept as the final one. This allows us to apply the matrix inversion approach as the first step of the iteration procedure suggested in this work. Thus, the final expression which explains how to interpret the

summation sign for the first step of the chosen procedure is

$$\sum_{\mathbf{k}} g(\mathbf{k}) = \frac{1}{4\pi^2} \sum_{j=0}^{N_k-1} W_j \int_0^{2\pi} d\varphi g(k_j, \varphi), \quad (\text{D4})$$

where weight functions W_i and points k_i depend on the partition of the interval $[0, k_{\max}]$. In order to achieve the better convergence we choose the points of the Gauss-Legendre quadrature support. Taking into account the interesting interval the corresponding functions can be expressed as follows:

$$k_i = (Z_{L,i} + 1) \frac{k_{\max}}{2}, \quad W_i = k_i W_{L,i} \frac{k_{\max}}{2}, \quad (\text{D5})$$

where $Z_{L,i}$ is the i th root of the Legendre polynomial P_{N_k} , while weights are given by the formula

$$W_{L,i} = \frac{2}{(1 - Z_{L,i}^2) [P'_{N_k}(Z_{L,i})]^2}. \quad (\text{D6})$$

With these expressions in hand the system of equations (40) can be rewritten as follows:

$$\sum_{j=0}^{N_k-1} M_{i,j} \Gamma_{II,j} = 1, \quad \Gamma_{II,j} \equiv \Gamma_{II}(k_j), \quad \text{for } i = 0, \dots, N_k - 1, \quad (\text{D7})$$

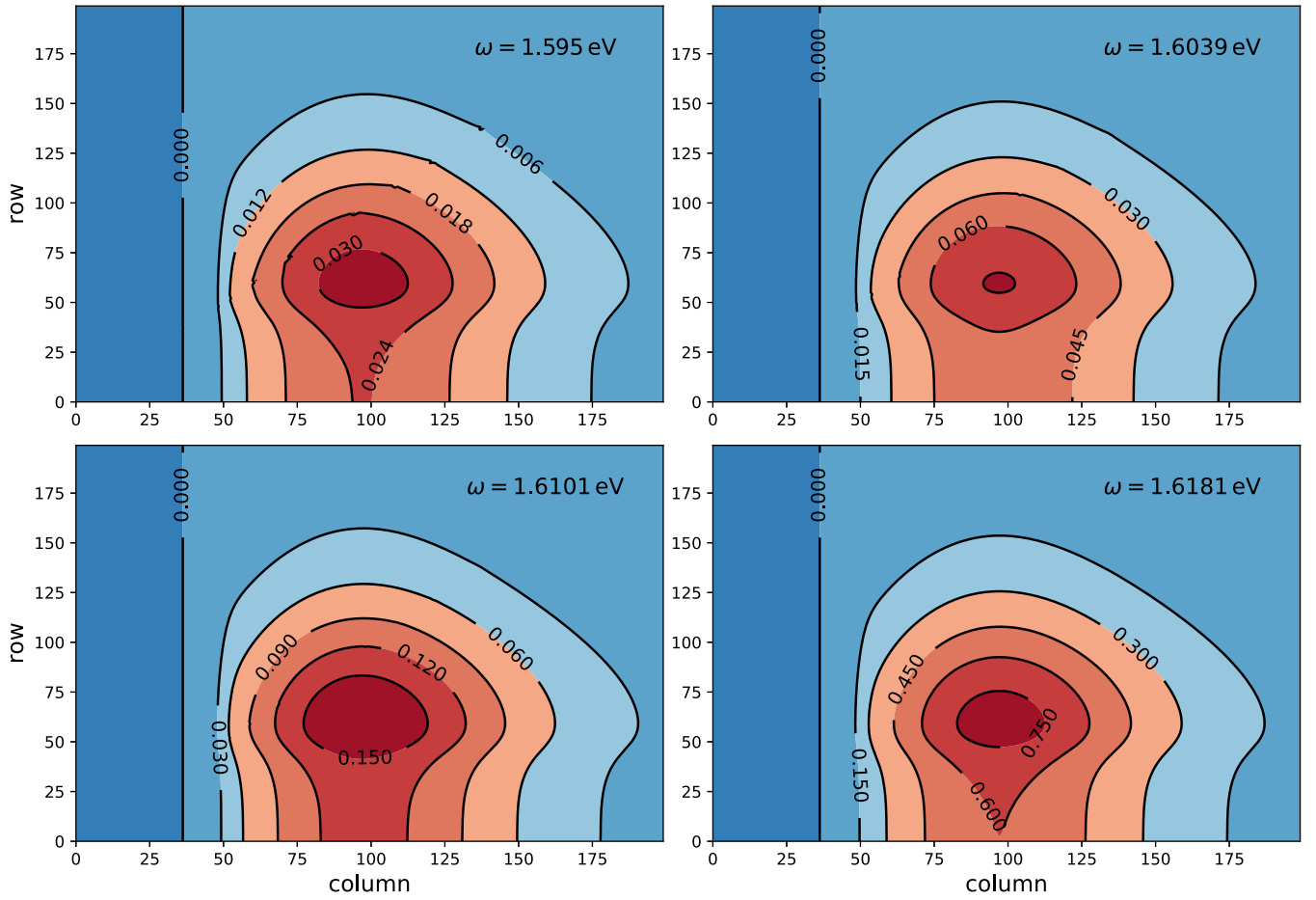


FIG. 21. The behavior of imaginary part of matrix coefficients $\hat{M}_{i,j}^{-1}$ for different values of frequency ω . This matrix was used to calculate the absorption in Fig. 10. The values of physical parameters including temperature and carrier densities can be found also there.

$$M_{i,j} = \delta_{i,j} + \frac{(\delta_{i,j} - 1)}{d_{cv}} \frac{1}{4\pi^2} W_j \chi_I^R(k_j, \omega) \times \int_0^{2\pi} d\varphi V(k_i^2 + k_j^2 + 2k_i k_j \cos \varphi), \quad (\text{D8})$$

where d_{cv} is effective dipole moment, which we have taken equal to 1 nm. The matrix obtained can be reversed. This

allows one to find all $\Gamma_{II,j}$ as

$$\vec{\Gamma}_{II} = \hat{M}^{-1} \vec{E}_{II}, \quad \vec{E}_{II} = (1, \dots, 1). \quad (\text{D9})$$

Having obtained the solution for Γ_{II} , we can easily find the value of $\chi_{II}(\omega)$ as

$$\chi_{II}(\omega) = \frac{d_{cv}}{2\pi} \sum_{j=0}^{N_k-1} \chi_2(k_j, \omega) = \frac{d_{cv}}{2\pi} \sum_{j=0}^{N_k-1} \Gamma_{2,j} \chi_I^R(k_j, \omega). \quad (\text{D10})$$

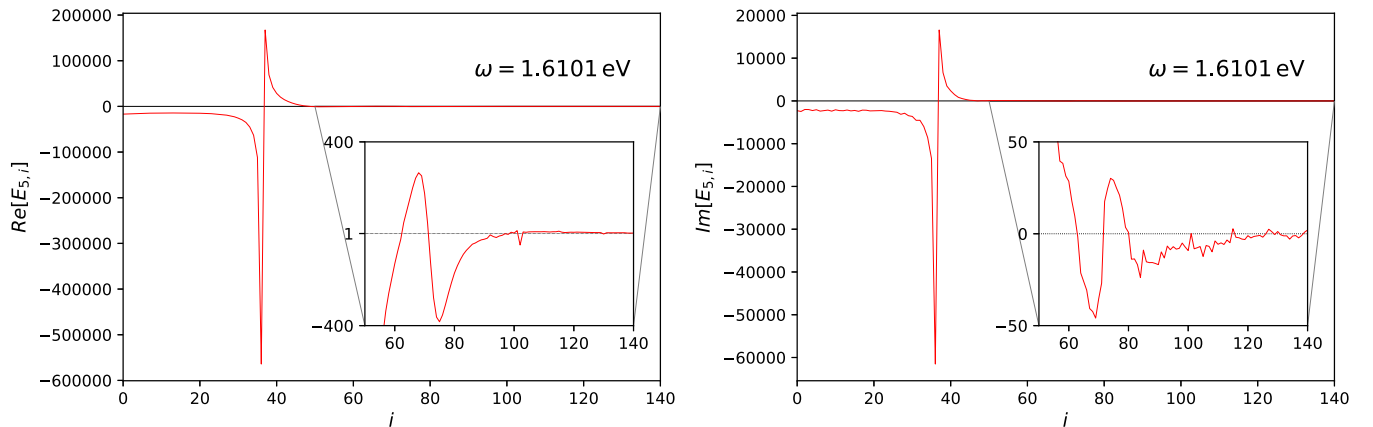


FIG. 22. The behavior of imaginary and real parts of $E_{V,i}$ elements.

Depending on the charge carrier density the convergence is observed when N_k is varying from 300 to 600. The obtained points $\chi_2(k_j, \omega)$ are stored for the needs of the further calculations.

2. Beyond the Bloch approximation

In order to obtain the correction (C68) for (C69), we have to perform the double summation, which includes essentially $\chi_{II}(\mathbf{k}, \omega)$ from the previous step. Instead of direct summation we compute the four-dimensional integrals by means of the Monte Carlo technique. This choice is dictated by the following reason. The extremely cumbersome integrand leads to the difficulties with analysis of the problematic regions in momentum space where we have to place a greater focus. The replacement of summation by the adaptive Monte Carlo

scheme allows us to overcome this problem and often reduce the number of points in order to achieve the required accuracy. However, as can be noted, for performing of the integration one has to know the continuous χ dependence on momentum over the entire interval, not only in the separate points. For this purpose we construct the piecewise linear interpolation which gives the opportunity to highly accurately reconstruct the $\chi_{II}(\omega)$ behavior with many fewer points (the value of N_k) than was necessary for the initial calculations when the matrix inversion method is used (the reduction from 600 to 60 points in momentum space for a specific value of ω).

Returning to the calculation of χ_{III} , first, we have to calculate (C68). The result of calculations is used for (C70) and consequently for (C69). In terms of Γ one has to solve the following system of equations which is modified as compared with (D7):

$$\sum_{j=0}^{N_k-1} M_{i,j} \Gamma_{III,j} = 1 + F_{\Gamma}(\mathfrak{D}^{\omega,III}, \mathbf{k}_i, \omega), \quad \Gamma_{III,j} \equiv \Gamma_{III}(k_j), \quad \text{for } i = 0, \dots, N_k - 1, \quad (\text{D11})$$

$$F_{\Gamma}(\mathfrak{D}^{\omega}, \mathbf{k}, \omega) = -\frac{1}{\mathcal{E}(\omega) d_{cv} [1 - f_{(e,\mathbf{k})} - f_{(h,\mathbf{k})}]} \sum_{\mathbf{k}'_2, \mathbf{q}' \neq 0} V_{\mathbf{q}'} [\mathfrak{D}^{\omega}_{(c,\mathbf{k}-\mathbf{q}'),(c,\mathbf{k}'_2),(c,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k})} + \mathfrak{D}^{\omega}_{(c,\mathbf{k}-\mathbf{q}'),(v,\mathbf{k}'_2),(v,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k})} - \mathfrak{D}^{\omega}_{(c,\mathbf{k}),(v,\mathbf{k}'_2),(v,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k}+\mathbf{q}')} - \mathfrak{D}^{\omega}_{(c,\mathbf{k}),(c,\mathbf{k}'_2),(c,\mathbf{k}'_2-\mathbf{q}'),(v,\mathbf{k}+\mathbf{q}')}], \quad (\text{D12})$$

with the same $M_{i,j}$ from (D8). The new vector $\vec{\Gamma}_{III}$ can be found by means of an equation similar to (D9):

$$\vec{\Gamma}_{III} = \hat{M}^{-1} \vec{E}_{III}, \quad E_{III,i} = 1 + F_{\Gamma}(\mathfrak{D}^{\omega,III}, \mathbf{k}_i, \omega), \quad \text{for } i = 0, \dots, N_k - 1. \quad (\text{D13})$$

As is easily seen, such equation can be generalized for $N = IV, V$:

$$\vec{\Gamma}_N = \hat{M}^{-1} \vec{E}_N, \quad E_{N,i} = 1 + F_{\Gamma}(\mathfrak{D}^{\omega,N}, \mathbf{k}_i, \omega), \quad \text{for } i = 0, \dots, N_k - 1, \quad \text{and } N = III, IV, V. \quad (\text{D14})$$

Thus, in order to obtain one point in ω space, we need to calculate all $E_{N,i}$. In this regard, it would be reasonable to analyze the behavior of matrix \hat{M} . The behavior of real and imaginary parts of $\hat{M}_{i,j}^{-1}$ is presented in Figs. 20 and 21, respectively. First, the relative structure of the matrix element behavior almost does not depend on the frequency value; the maximum value is located within the unchanged vicinity. Second, these figures allow us to recognize which components of $E_{N,i}$ are of prime importance. Speaking about the vector \vec{E}_N itself, we present the typical behavior of its real and imaginary parts for the most computationally expensive case: $N = V$. These dependencies are shown in Fig. 22. Having analyzed both objects— \hat{M}^{-1} and \vec{E} —it is easy to conclude that there is no need to calculate all the points $i = 0, \dots, N_k - 1$ in momentum space. In particular, the most expensive approximation from the computation point of view— $N = V$ —demands only the knowledge of $E_{V,i}$ for i no higher than ≈ 140 . We also exclude (replace $E_{N,i}$ by pure unity) the contributions i , the relative accuracy of which is worse than 0.4. The number of such contributions is always around 10% and most of them are sitting in the tail. Taking them into account moderately affects only the height of the trionic peak, leaving, however, its position unchanged. This fact—the partial accounting of $E_{N,i}$ contributions—crucially decreased the computational time. In the case of $N = V$ (eight-dimensional integration) the number of Monte Carlo evaluations for each couple (k_i, ω_j) is 1.5×10^9 . In such a dramatic situation we address the GPU opportunities and gVEGAS—the GPU implementation of the well-known Monte Carlo algorithm [57].

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