

Quantum kinetic description of spin transfer in diluted magnetic semiconductors

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We derive quantum kinetic equations of motion that describe laser-driven Mn-doped diluted semiconductors and account for the carrier Mn exchange interaction beyond the mean-field theory. We treat a spatially inhomogeneous system with arbitrary given positions of Mn dopants as well as an ensemble of randomly distributed Mn atoms in an infinite crystal, which represents an on average spatially homogeneous system. In the latter case, special care is taken of the interplay between higher-order correlations and the random positioning of Mn atoms. For the ensemble-averaged system, we explicitly identify the terms responsible for a spin transfer between spin-polarized carriers and Mn atoms in the special case valid, e.g., for paramagnetic samples without external magnetic field, where initially the total Mn magnetization vanishes. It turns out that here the mean-field approach as well as the virtual crystal approximation predict a vanishing spin transfer, in contrast to our quantum kinetic equations. Moreover, in our approach, the exchange interaction with the localized Mn atoms leads to a significant redistribution of the carrier momenta even in an on average spatially homogeneous system. The latter feature can not be described in the virtual crystal approximation.

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I. INTRODUCTION

Diluted magnetic semiconductors (DMS) keep being a focus of intensive research.¹⁻⁴ On the one hand, the possibility to integrate magnetic properties in a semiconductor environment is of high technological interest and, on the other hand, these materials give rise to many open questions of physical relevance. DMS (mostly Mn-doped semiconductors) are particularly often discussed as a hardware for spintronic applications. For these purposes, a targeted control of the dynamics of carrier and Mn spins in the material is of utmost importance. Such a control can be achieved via electrical currents (transport of spins)^{5,6} or via optical manipulation of the carriers (spin orientation).⁷⁻⁹ Optical control usually can be realized on a much shorter time scale than control by transport, but one has to face the challenge that the direct coupling of the laser field to the Mn spins is negligible. Thus, a control can only be achieved indirectly via the spin transfer between optically excited carriers and the Mn system mediated by the exchange interaction.

While the number of DMS with different compositions steadily increases, by far the most studied systems are Mn-doped II-VI and III-V semiconductors.^{10,11} In both cases, the Mn atoms effectively provide localized spins $5/2$ due to their half-filled d shell. Moreover, the localized spins interact with the spins of itinerant electrons and holes via an exchange mechanism. III-V-semiconductor-based DMS are ferromagnetic when the Mn concentration is above a certain threshold (typically 2%).¹² Here, the Mn atoms normally act as acceptors leading to itinerant hole concentrations of up to 10^{20} cm^{-3} . The ferromagnetic ordering of the Mn spins is caused by the Mn carrier exchange interaction mediated via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism. In contrast, in II-IV materials, the Mn atoms mostly substitute the host atoms of group II isoelectronically and therefore no free carriers are available that could mediate the RKKY interaction. In these materials, the Mn doping typically leads to a rather strong paramagnetism.

The modeling of the dynamics in DMS is mostly done either on the level of rate equations¹³⁻¹⁵ or by treating the carrier Mn exchange interaction on the mean-field level, typically in combination with the virtual crystal approximation.^{16,17} Rate equations describe the spin transfer between carriers and Mn system as an incoherent process and thus miss coherent features that, e.g., enable coherent control of the Mn spin dynamics.¹⁸⁻²¹ The mean-field approach yields such a spin transfer only when the initial average Mn magnetization is nonzero and is commonly used to describe Mn spin precession in ferromagnets or paramagnets with an external magnetic field. The virtual crystal approximation disregards that the Mn atoms are localized at certain positions. Recently, for the ferromagnetic case, corrections to the mean-field theory have been worked out within the Green's function formalism.^{22,23} In the latter works, a spatially inhomogeneous situation has been considered and, consequently, the virtual crystal approximation has not been used.

In this paper, we develop a microscopic density matrix approach to the dynamics of DMS, which accounts for exchange-induced correlations beyond the mean-field level and the localized character of the Mn spins. Our theory can be applied to both ferromagnetic and paramagnetic DMS. Within the density matrix theory, the crucial step to obtain a numerically tractable set of equations is to truncate the hierarchy of equations of motion for higher-order correlation functions. We present a systematic truncation scheme that can be applied to a system with arbitrary given positions of the Mn atoms. The scheme fully accounts for density matrices involving more than one spin operator for a given Mn site by using exact relations that allow a reduction to density matrices with only one spin operator per site. Density matrices with spin operators referring to different Mn atoms are treated within a correlation expansion. In addition, we also derive a description for systems that are spatially homogeneous after averaging over a uniform distribution of Mn atom positions. The simplifications that result from the assumption of a spatially homogeneous Mn distribution are

worked out in this paper explicitly for bulk semiconductors; analogous equations for quantum wells or quantum wires that are on average translational invariant in the direction parallel to the well or wire can be obtained easily with one additional approximation that is explained in Appendix B. It turns out that the combination of accounting for higher-order correlations with the averaging procedure yields results that deviate from the virtual crystal predictions even in an on average spatially homogeneous system. We explicitly discuss the special case of a spatially homogeneous system where initially the Mn magnetization is zero and identify the contributions in our equations that are responsible for a spin transfer from initially spin-polarized carriers toward the Mn atoms under these conditions. The elementary step in a spin-transfer process that is mediated by the exchange interaction is a change of a carrier spin by ± 1 that is accompanied by a change of the spin of a Mn atom by ∓ 1 . Consequently, starting from a full spin-polarized carrier distribution, the only possible process is a decrease of a carrier spin accompanied by an increase of the spin of a Mn atom. Thus, an average Mn magnetization that is initially zero must acquire finite values in the course of time as is also already seen in early experiments.^{15,24} Nevertheless, our analysis reveals that this spin transfer is absent not only within the mean-field theory, but also in the limit of an infinite crystal within the virtual crystal approach. Furthermore, we show that such a spin transfer is accompanied by a significant redistribution of the carrier momenta even if the system is spatially homogeneous on average. This feature is missing completely within the virtual crystal approximation. It turns out that it is crucial to account properly for the interplay between spatial averaging and the dynamics of higher-order correlations.

II. MODEL SYSTEM

The starting point for our analysis is the Hamiltonian

$$H = H_0 + H_{sd}, \quad (1)$$

which consists of the single-particle electron part H_0 and the s - d exchange interaction H_{sd} between s -like conduction-band electrons and localized Mn spins.

In the main part of this paper, we do not account for the exchange interaction H_{pd} between p -like holes in the valence bands and the Mn atoms since all principal conclusions of this paper can be drawn readily and more conveniently from the reduced model $H_0 + H_{sd}$. An extended set of equations for spatially homogeneous systems in the large-volume limit that accounts for H_{pd} is given in Appendix C, where we also work out the inclusion of a coupling to an external laser field that is needed for the optical excitation of spin-polarized carriers. In the main part, the optical excitation is modeled by appropriate initial conditions.

We consider electrons in two spin-degenerate conduction bands with spin quantum number $\frac{1}{2}$. Since we assume that the bands are purely s like, we neglect band-mixing effects. With these assumptions, H_0 reads as

$$H_0 = \sum_{\sigma\mathbf{k}} E_{\mathbf{k}} c_{\sigma\mathbf{k}}^\dagger c_{\sigma\mathbf{k}}, \quad (2)$$

where $\sigma \in \{\downarrow\uparrow\}$ is the spin index, \mathbf{k} the three-dimensional wave vector, and $E_{\mathbf{k}}$ the corresponding energy.

The exchange interaction, which originates from hybridization effects between s -like conduction-band states and d -like Mn orbitals,^{25,26} is described by a Kondo-type Hamiltonian

$$H_{sd} = J_{sd} \sum_{Ii} \hat{\mathbf{S}}_I \cdot \hat{\mathbf{s}}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I), \quad (3)$$

where $\hat{\mathbf{S}}_I$ ($\hat{\mathbf{s}}_i^e$) is the spin operator and \mathbf{R}_I (\mathbf{r}_i) the position vector of the I th Mn atom (i th conduction-band electron). J_{sd} , which is also denoted by α in the literature, is the exchange constant with typical values of 10 meV nm³ in both (II, Mn)VI and (III, Mn)V semiconductors.^{13,27} The localized character of this interaction is reflected by the delta functions $\delta(\mathbf{r}_i - \mathbf{R}_I)$. For reasons which will become apparent later, we express $\hat{\mathbf{S}}_I$ in the basis $|I, n\rangle$ of eigenfunctions of its z component:

$$\hat{\mathbf{S}}_I = \sum_{nn'} \mathbf{S}_{nn'} \hat{P}_{nn'}^I, \quad (4)$$

where $n \in \{-\frac{5}{2}, -\frac{3}{2}, \dots, \frac{5}{2}\}$ is the magnetic spin quantum number, $\mathbf{S}_{nn'} := \langle I, n | \hat{\mathbf{S}}_I | I, n' \rangle$ are the spin matrix elements, and

$$\hat{P}_{nn'}^I := |I, n\rangle \langle I, n'| \quad (5)$$

are the new operators for the Mn spin degrees of freedom. In the second quantization, Eq. (3) becomes

$$H_{sd} = \frac{J_{sd}}{V} \sum_{\substack{I, n, n' \\ \sigma, \sigma', \mathbf{k}, \mathbf{k}'}} \mathbf{S}_{nn'} \cdot \mathbf{s}_{\sigma\sigma'}^e e^{i(\mathbf{k}' - \mathbf{k})\mathbf{R}_I} c_{\sigma\mathbf{k}}^\dagger c_{\sigma'\mathbf{k}'} \hat{P}_{nn'}^I, \quad (6)$$

where V is the volume of the sample and $\mathbf{s}_{\sigma\sigma'}^e$ is the vector of Pauli matrices.

III. EQUATIONS OF MOTION

In this section, we derive two sets of equations of motion describing the combined electron and Mn dynamics. The first set of equations deals with the case of known and fixed positions of the Mn atoms, while the second is derived based on the assumption of a spatially homogeneous random distribution of Mn atoms in a macroscopic system, i.e., we consider the limit of an infinite system volume ($V \rightarrow \infty$) and number of Mn atoms ($N_{\text{Mn}} \rightarrow \infty$) where the Mn density n_{Mn} is kept finite. Altogether, the second case corresponds to an average spatially homogeneous system.

A. Equations for fixed Mn positions

1. Heisenberg equations

The first step of our derivation is to set up the Heisenberg equations of motion for the expectation values of the Mn operators

$$\begin{aligned} -i\hbar \frac{\partial}{\partial t} \langle \hat{P}_{n_1 n_2}^I \rangle &= \frac{J_{sd}}{V} \sum_{\substack{n\sigma\sigma' \\ \mathbf{k}\mathbf{k}'}} (\mathbf{S}_{nn_1} \langle c_{\sigma\mathbf{k}}^\dagger c_{\sigma'\mathbf{k}'} \hat{P}_{n_2}^I \rangle \\ &\quad - \mathbf{S}_{n_2 n} \langle c_{\sigma\mathbf{k}}^\dagger c_{\sigma'\mathbf{k}'} \hat{P}_{n_1}^I \rangle) \cdot \mathbf{s}_{\sigma\sigma'}^e e^{i(\mathbf{k}' - \mathbf{k})\mathbf{R}_I}, \end{aligned} \quad (7a)$$

and for the electronic two-point density matrices

$$\begin{aligned} & \left(-i\hbar \frac{\partial}{\partial t} + E_{\mathbf{k}_2} - E_{\mathbf{k}_1} \right) \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \\ &= \frac{J_{sd}}{V} \sum_{I'} \sum_{\substack{nn' \\ \sigma \mathbf{k}}} \mathbf{S}_{nn'} \cdot (\mathbf{s}_{\sigma \sigma_1}^e \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{nn'}^{I'} \rangle e^{i(\mathbf{k}_1 - \mathbf{k})\mathbf{R}_{I'}} \\ & \quad - \mathbf{s}_{\sigma_2 \sigma}^e \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}} \hat{P}_{nn'}^I \rangle e^{i(\mathbf{k} - \mathbf{k}_2)\mathbf{R}_{I'}}). \end{aligned} \quad (7b)$$

Due to the exchange interaction, these variables are coupled to three-point density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle$, which describe correlations between Mn atoms and electrons, but still contain mean-field contributions $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle \hat{P}_{n_1 n_2}^I \rangle$. In order to separate these mean-field contributions from true correlations between electrons and Mn atoms, we introduce

$$\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle := \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle \hat{P}_{n_1 n_2}^I \rangle,$$

i.e., the deviations from the mean-field factorizations, as new dynamical variables instead of $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle$. The equations of motion for these true correlations read as

$$\begin{aligned} & \left(-i\hbar \frac{\partial}{\partial t} + E_{\mathbf{k}_2} - E_{\mathbf{k}_1} \right) \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle \\ &= Q_{\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle}^{\text{III}} + Q_{\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle}^{\text{IV}} + Q_{\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle}^{\text{V}}. \end{aligned} \quad (7c)$$

In the above equations, the source terms

$$\begin{aligned} Q_{\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle}^{\text{III}} &:= \frac{J_{sd}}{V} \sum_{n\sigma \mathbf{k}} [\mathbf{S}_{nn_1} \cdot \mathbf{s}_{\sigma \sigma_1}^e e^{i(\mathbf{k}_1 - \mathbf{k})\mathbf{R}_I} \\ & \quad \times (\delta \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{nn_2}^I \rangle + \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle \hat{P}_{nn_2}^I \rangle) \\ & \quad - \mathbf{S}_{n_2 n} \cdot \mathbf{s}_{\sigma_2 \sigma}^e e^{i(\mathbf{k} - \mathbf{k}_2)\mathbf{R}_I} (\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}} \hat{P}_{n_1 n}^I \rangle \\ & \quad + \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}} \rangle \langle \hat{P}_{n_1 n}^I \rangle)] \end{aligned} \quad (8a)$$

collect contributions from three-point functions and their factorized counterparts, while

$$\begin{aligned} Q_{\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle}^{\text{IV}} &:= \frac{J_{sd}}{V} \sum_{I' \neq I} \sum_{\substack{nn' \\ \sigma \mathbf{k}}} \mathbf{S}_{nn'} \cdot (\mathbf{s}_{\sigma \sigma_1}^e e^{i(\mathbf{k}_1 - \mathbf{k})\mathbf{R}_{I'}} \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \hat{P}_{nn'}^{I'} \rangle \\ & \quad - \mathbf{s}_{\sigma_2 \sigma}^e e^{i(\mathbf{k} - \mathbf{k}_2)\mathbf{R}_{I'}} \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}} \hat{P}_{n_1 n_2}^I \hat{P}_{nn'}^{I'} \rangle) \\ & \quad - \langle \hat{P}_{n_1 n_2}^I \rangle \frac{J_{sd}}{V} \sum_{I'} \sum_{\substack{nn' \\ \sigma \mathbf{k}}} \mathbf{S}_{nn'} \cdot (\mathbf{s}_{\sigma \sigma_1}^e e^{i(\mathbf{k}_1 - \mathbf{k})\mathbf{R}_{I'}} \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{nn'}^{I'} \rangle \\ & \quad - \mathbf{s}_{\sigma_2 \sigma}^e e^{i(\mathbf{k} - \mathbf{k}_2)\mathbf{R}_{I'}} \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}} \hat{P}_{nn'}^{I'} \rangle), \end{aligned} \quad (8b)$$

$$\begin{aligned} Q_{\delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle}^{\text{V}} &:= -\frac{J_{sd}}{V} \sum_{\substack{n\sigma \sigma' \\ \mathbf{k} \mathbf{k}'}} (\mathbf{S}_{nn_1} \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} c_{\sigma' \mathbf{k}'} \hat{P}_{nn_2}^I \rangle \\ & \quad - \mathbf{S}_{n_2 n} \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} c_{\sigma' \mathbf{k}'} \hat{P}_{n_1 n}^I \rangle) \cdot \mathbf{s}_{\sigma \sigma'}^e e^{i(\mathbf{k}' - \mathbf{k})\mathbf{R}_I} \end{aligned}$$

$$\begin{aligned} & - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \frac{J_{sd}}{V} \sum_{\substack{n\sigma \sigma' \\ \mathbf{k} \mathbf{k}'}} (\mathbf{S}_{nn_1} \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma' \mathbf{k}'} \hat{P}_{nn_2}^I \rangle \\ & \quad - \mathbf{S}_{n_2 n} \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma' \mathbf{k}'} \hat{P}_{n_1 n}^I \rangle) \cdot \mathbf{s}_{\sigma \sigma'}^e e^{i(\mathbf{k}' - \mathbf{k})\mathbf{R}_I} \end{aligned} \quad (8c)$$

collect the contributions of higher density matrices, i.e., four-point density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \hat{P}_{n_3 n_4}^{I'} \rangle$ with $I' \neq I$ and normal ordered five-point density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} \hat{P}_{n_1 n_2}^I \rangle$ and corresponding factorizations.

2. One-Mn-site density matrices

The restriction $I' \neq I$ on the Mn indices in Eq. (8b) is due to the fact that density matrices with $I' = I$, which we refer to as *one-Mn-site density matrices*, can be reduced to three-point functions without approximation according to

$$\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \hat{P}_{n_3 n_4}^I \rangle = \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_4}^I \rangle \delta_{n_2 n_3}. \quad (9)$$

Here, we have used the identity

$$\hat{P}_{n_1 n_2}^I \hat{P}_{n_3 n_4}^I = \hat{P}_{n_1 n_4}^I \delta_{n_2 n_3}, \quad (10)$$

which follows directly from the definition of the operators in Eq. (5). The term Q^{III} on the right-hand side of Eq. (7c) arises from both the above relation and the normal ordering of the five-point functions. As will be shown later, this term is decisive for the spin-transfer mechanism discussed in Sec. IV.

3. Truncation scheme

The higher-order density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \hat{P}_{n_3 n_4}^{I'} \rangle$ with $I' \neq I$, which we denote as *two-Mn-site density matrices*, and the density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} \hat{P}_{n_1 n_2}^I \rangle$ that involve four electron operators lead to an infinite hierarchy of equations of motion, which is not tractable without further approximation.

In order to truncate this hierarchy, we apply a standard correlation expansion²⁸ and decompose all four- and five-point density matrices in Eq. (7c) into sums of all possible products of the basic irreducible functions $\langle \hat{P}_{n_1 n_2}^I \rangle$ and $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle$ and the following irreducible rests:

$$\begin{aligned} & \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle, \\ & \delta \langle P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle := \langle P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle - \langle P_{n_1 n_2}^{I'} \rangle \langle P_{n_3 n_4}^I \rangle, \\ & \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} \rangle \\ & \quad := \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} \rangle - (\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_4 \mathbf{k}_4} \rangle \langle c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} \rangle \\ & \quad \quad - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_3 \mathbf{k}_3} \rangle \langle c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_4 \mathbf{k}_4} \rangle), \\ & \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle, \quad \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} P_{n_1 n_2}^I \rangle. \end{aligned}$$

The last two functions are implicitly defined by the rather lengthy decompositions of the functions $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle$ and $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} P_{n_1 n_2}^I \rangle$ in irreducible parts, which is given explicitly in Eqs. (A1) and (A2). All five irreducible rests describe true higher-order correlations, i.e., the deviation of the corresponding density matrix from its factorized form.

In this paper, we select relevant dynamical variables according to the following arguments. Correlations which contain Mn operators belonging to different atoms are expected to be small since their sites are typically far apart in diluted

magnetic semiconductors. Thus, we can neglect $\delta\langle P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle$ and $\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle$. We shall also assume that correlations involving four electronic operators are of minor importance, i.e., we also disregard $\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} \rangle$ and $\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}^\dagger c_{\sigma_3 \mathbf{k}_3} c_{\sigma_4 \mathbf{k}_4} P_{n_1 n_2}^I \rangle$. This reduction of dynamical variables results in a closed set of equations of motion for the remaining quantities $\langle \hat{P}_{n_1 n_2}^I \rangle$, $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle$, and $\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle$. In this description, the mean-field limit is recovered when the correlations $\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle$ are neglected. However, these correlations are the driving force for important parts of the dynamics. For instance, the spin transfer from spin-polarized carriers to Mn atoms starting from an initially zero Mn magnetization discussed in Sec. IV could not be described without $\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle$ (cf. Sec. V).

We also would like to stress that it is of utmost importance to use the above correlation expansion only for the two-Mn-site density matrices with $I' \neq I$. For one-Mn-site density matrices, such an expansion is not necessary as these quantities do not lead to a hierarchy of equations of motion, but can rather be exactly expressed by density matrices involving only a single Mn operator according to Eq. (9). Applying the same factorization that is used here for two-Mn-site density matrices also to the one-Mn-site density matrices may lead to severe errors, e.g., in the description of the spin-transfer mechanism in Sec. IV.

We note in passing that for the separate treatment of one-Mn-site and two-Mn-site density matrices, it was rather advantageous to represent the Mn spin degrees of freedom by the operators $\hat{P}_{n_1 n_2}^I$ and not by the spin operators \hat{S}_I that are common in the literature. In a representation based on spin operators, one would have to deal with one-Mn-site density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} (\hat{S}_I)^j \rangle$, which contain arbitrary powers $j \in \mathbb{N}$ of \hat{S}_I . Using the spin algebra of \hat{S}_I , it is much more cumbersome to exactly reduce these variables to a finite closed set than in a representation based on the operators $\hat{P}_{n_1 n_2}^I$, where the simple relation (10) can be used.

B. Spatially homogeneous disorder-averaged dynamics

The precise positions of the Mn atoms are usually not known, and typical experiments measure effectively an average over a more or less random distribution of these positions. Such disorder effects can be accounted for by introducing density matrices that are averaged over the distribution of Mn positions, e.g., when writing

$$\langle \hat{P}_{n_1 n_2}^I \rangle, \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle, \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle,$$

the brackets $\langle \dots \rangle$ from now on imply that after taking the quantum mechanical expectation value for a fixed configuration of Mn positions, the result is additionally averaged over a random distribution of Mn positions. By performing the additional disorder averaging in Eqs. (7a) and (7b), one obtains equations for the now disorder-averaged variables $\langle \hat{P}_{n_1 n_2}^I \rangle$ and $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle$, which differ from Eqs. (7a) and (7b) only by the fact that the phase factors that involve the positions \mathbf{R}_I can not be taken out of the expectation value any more. Thus, e.g.,

Eq. (7b) becomes

$$\begin{aligned} & \left(-i\hbar \frac{\partial}{\partial t} + E_{\mathbf{k}_2} - E_{\mathbf{k}_1} \right) \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \\ &= \frac{J_{sd}}{V} \sum_{I'} \sum_{\substack{nn' \\ \sigma \mathbf{k}}} \mathbf{S}_{nn'} \cdot \left(s_{\sigma \sigma_1}^e \langle c_{\sigma \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{nn'}^{I'} e^{i(\mathbf{k}_1 - \mathbf{k}) \mathbf{R}_{I'}} \rangle \right. \\ & \quad \left. - s_{\sigma_2 \sigma}^e \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma \mathbf{k}} \hat{P}_{nn'}^{I'} e^{i(\mathbf{k} - \mathbf{k}_2) \mathbf{R}_{I'}} \rangle \right), \end{aligned} \quad (11)$$

which contains the new function

$$\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle. \quad (12)$$

The corresponding hierarchy of equations of motion can again be reformulated in terms of correlation functions where, different from the previously discussed case, the true correlations are defined with respect to disorder-averaged variables. It should be noted that phase factors, such as $e^{i\mathbf{k}_3 \mathbf{R}_I}$ in Eq. (12), are for $\mathbf{k}_3 \neq 0$ random variables due to their dependence on \mathbf{R}_I and thus must be treated in the correlation expansion²⁸ analogously to the operators $c^\dagger c$ or \hat{P} . For example, the function in Eq. (12) involves for $\mathbf{k}_3 = 0$ an average over the operators $c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2}$ and $\hat{P}_{n_1 n_2}^I$, where the correlated part can be defined as

$$\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle := \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle \hat{P}_{n_1 n_2}^I \rangle. \quad (13a)$$

For $\mathbf{k}_3 \neq 0$, the phase $e^{i\mathbf{k}_3 \mathbf{R}_I}$ appears as an additional factor that has to be averaged and thus the decomposition into factorized and correlated parts reads as

$$\begin{aligned} & \delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle \\ &:= \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle \hat{P}_{n_1 n_2}^I \rangle \langle e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle \\ & \quad - \delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle \langle e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle - \delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle \langle \hat{P}_{n_1 n_2}^I \rangle \\ & \quad - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \delta\langle \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle, \end{aligned} \quad (13b)$$

where two additional correlation functions appear, namely,

$$\delta\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle := \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle - \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle \quad (13c)$$

and

$$\delta\langle \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle := \langle \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle - \langle \hat{P}_{n_1 n_2}^I \rangle \langle e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle. \quad (13d)$$

From the equations for the disorder-averaged density matrices, it is straightforward to derive equations of motion for the correlation functions defined by Eqs. (13). These equations involve the disorder-averaged higher-order density matrices

$$\begin{aligned} & \langle c_i^\dagger c_j^\dagger c_k c_l \hat{P}^I e^{i\mathbf{k} \mathbf{R}_I} \rangle, \quad \langle c_i^\dagger c_j^\dagger \hat{P}^I \hat{P}^{I'} e^{i\mathbf{k} \mathbf{R}_I} e^{i\mathbf{k}' \mathbf{R}_{I'}} \rangle, \\ & \langle c_i^\dagger c_j^\dagger \hat{P}^I \hat{P}^{I'} e^{i\mathbf{k} \mathbf{R}_I} \rangle, \quad \langle c_i^\dagger c_j^\dagger \hat{P}^I e^{i\mathbf{k} \mathbf{R}_I} e^{i\mathbf{k}' \mathbf{R}_{I'}} \rangle, \langle c_i^\dagger c_j^\dagger \hat{P}^I e^{i\mathbf{k} \mathbf{R}_I} \rangle. \end{aligned}$$

In order to obtain a closed set of equations, these density matrices can be decomposed into factorized parts involving only lower-order correlation functions and a remaining high-order correlation function. Neglecting the latter results in a finite set of coupled dynamical variables which is, however, in the general case of an arbitrary random distribution of Mn

positions way too complicated for practical applications and is therefore not given here explicitly.

Following the arguments discussed in connection with the truncation scheme for the case without disorder averaging (cf. Sec. III A3), we restrict the correlation functions that are kept to the dynamical variables defined in Eqs. (13). In addition, we concentrate on the description of diluted systems where the Mn atoms are distributed independently and homogeneously in space, i.e., we assume a random distribution of Mn atoms where on average no point in space is distinguished and derive the equations of motion relevant for this limit. A spatially homogeneous description is often a good approximation for bulk DMS.¹² Obvious requirements for reaching the spatially homogeneous limit is that only external fields with negligible spatial variations may be considered, and the effects related to the aggregation of Mn clusters²⁹ may be disregarded. Although for samples where translational invariance holds only in one or two dimensions, such as quantum wires or quantum wells, the assumption of a fully spatially homogeneous system does not strictly apply, simplified equations can be derived that are similar to the three-dimensional homogeneous equations. The necessary modifications for these cases are presented in Appendix B.

Assuming a spatially uniform random distribution of Mn positions, the average $\langle e^{i\mathbf{k}\mathbf{R}_I} \rangle$ can be easily performed, resulting in

$$\langle e^{i\mathbf{k}\mathbf{R}_I} \rangle = \delta_{\mathbf{k}\mathbf{0}}. \quad (14)$$

For a system where no point in space is distinguished, all directly observable quantities must be independent of position variables. Thus, our assumption of an on average spatially homogeneous system implies in particular that the values of single Mn variables

$$M_{n_1}^{n_2} := \langle \hat{P}_{n_1 n_2}^I \rangle \quad (15a)$$

do not depend on the index I . The same holds for all correlation functions defined in Eqs. (III B). Furthermore, the electronic density must be spatially homogeneous. Therefore, the electronic density matrix $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle$ must be diagonal in \mathbf{k} and thus only the variables

$$C_{\sigma_1 \mathbf{k}}^{\sigma_2} := \langle c_{\sigma_1 \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}} \rangle \quad (15b)$$

can have nonzero values. The requirement that the source terms for the off-diagonal components of $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle$ with $\mathbf{k}_1 \neq \mathbf{k}_2$ must vanish can in general be fulfilled only when $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle$ is nonzero only for $\mathbf{k}_3 = \mathbf{k}_2 - \mathbf{k}_1$ as can be seen by inspection of the corresponding equation of motion (11). By combining this requirement with the decomposition Eq. (13b), it is found that the relevant nonzero variables that have to be kept in the spatially homogeneous limit in addition to $M_{n_1}^{n_2}$ and $C_{\sigma_1 \mathbf{k}}^{\sigma_2}$ are

$$\bar{C}_{\sigma_1 \mathbf{k}_1}^{\sigma_2 \mathbf{k}_2} := V \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_I} \rangle, \quad (15c)$$

$$K_{\sigma_1 n_1 \mathbf{k}}^{\sigma_2 n_2} := V \delta \langle c_{\sigma_1 \mathbf{k}}^\dagger c_{\sigma_2 \mathbf{k}} \hat{P}_{n_1 n_2}^I \rangle, \quad (15d)$$

$$\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2} := V \delta \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_I} \rangle. \quad (15e)$$

\bar{C} is an electronic correlation that arises only due to the ensemble averaging over the Mn positions because, in an ensemble, the average over the product $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_I} \rangle$ does in general not equal the product of the averages $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_I} \rangle$. Note that the definition of \bar{C} does not involve Mn spin operators. In contrast, K is a correlation between electrons and Mn spins that is present already for fixed Mn positions, i.e., when only the quantum mechanical average is performed. Finally, \bar{K} describes correlations that arise partly due to the electron Mn spin correlations induced by the exchange interaction at a given site and partly due to the ensemble averaging over the Mn positions.

In the definition of the correlation functions in Eqs. (15c)–(15e), we have included the volume V as a prefactor to ensure that the so-defined functions approach finite values in the macroscopic limit. As the assumption of a spatially homogeneous system is most meaningful for macroscopic systems, we shall in the following only discuss the limit $V \rightarrow \infty$. Specializing the truncation scheme outlined above to the spatially homogeneous case yields indeed a closed set of equations for the functions defined in Eqs. (15). It turns out that in the macroscopic limit $V \rightarrow \infty$, the correlation $K_{\sigma_1 n_1 \mathbf{k}}^{\sigma_2 n_2}$ [cf. Eq. (15d)] does not couple back to the other dynamical variables and thus can be disregarded. The resulting equations for the remaining variables $M_{n_1}^{n_2}$, $C_{\sigma_1 \mathbf{k}}^{\sigma_2}$, $\bar{C}_{\sigma_1 \mathbf{k}_1}^{\sigma_2 \mathbf{k}_2}$, and $\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}$, which are the central formal development of this paper, are given by

$$\begin{aligned} -i\hbar \frac{\partial}{\partial t} M_{n_1}^{n_2} &= \frac{J_{sd}}{(2\pi)^3} \int_{\text{BZ}} \sum_{n\sigma\sigma'} \mathbf{s}_{\sigma\sigma'}^e \cdot \left\{ \left(C_{\sigma\mathbf{k}}^{\sigma'} + \int_{\text{BZ}} \frac{\bar{C}_{\sigma\mathbf{k}}^{\sigma'\mathbf{k}'}}{(2\pi)^3} d^3\mathbf{k}' \right) (\mathbf{S}_{nn_1} M_{n_1}^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) \right. \\ &\quad \left. + \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{\sigma n \mathbf{k}}^{\sigma' n_2 \mathbf{k}'} - \mathbf{S}_{n_2n} \bar{K}_{\sigma n_1 \mathbf{k}}^{\sigma' n \mathbf{k}'}) \frac{d^3\mathbf{k}'}{(2\pi)^3} \right\} d^3\mathbf{k}, \end{aligned} \quad (16a)$$

$$\begin{aligned} -i\hbar \frac{\partial}{\partial t} C_{\sigma_1 \mathbf{k}_1}^{\sigma_2} &= J_{sd} n_{\text{Mn}} \sum_{nn'\sigma} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} \left[\mathbf{s}_{\sigma\sigma_1}^e \left(C_{\sigma\mathbf{k}_1}^{\sigma_2} + \int_{\text{BZ}} \frac{\bar{C}_{\sigma\mathbf{k}}^{\sigma_2 \mathbf{k}_1}}{(2\pi)^3} d^3\mathbf{k} \right) - \mathbf{s}_{\sigma_2\sigma}^e \left(C_{\sigma_1 \mathbf{k}_1}^{\sigma} + \int_{\text{BZ}} \frac{\bar{C}_{\sigma_1 \mathbf{k}_1}^{\sigma \mathbf{k}}}{(2\pi)^3} d^3\mathbf{k} \right) \right] \right. \\ &\quad \left. + \int_{\text{BZ}} (\mathbf{s}_{\sigma\sigma_1}^e \bar{K}_{\sigma n \mathbf{k}}^{\sigma_2 n' \mathbf{k}_1} - \mathbf{s}_{\sigma_2\sigma}^e \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma n' \mathbf{k}}) \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}, \end{aligned} \quad (16b)$$

$$\left(-i\hbar\frac{\partial}{\partial t} + E_{\mathbf{k}_2} - E_{\mathbf{k}_1}\right)\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2} = J_{sd} \sum_{nn'\sigma} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} [s_{\sigma\sigma_1}^e (n_{Mn} \bar{C}_{\sigma\mathbf{k}_1}^{\sigma_2\mathbf{k}_2} + C_{\sigma\mathbf{k}_2}^{\sigma_2}) - s_{\sigma_2\sigma}^e (n_{Mn} \bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2} + C_{\sigma_1\mathbf{k}_1}^{\sigma})] \right. \\ \left. + \frac{1}{(2\pi)^3} \int_{\text{BZ}} [(s_{\sigma\sigma_1}^e \bar{K}_{\sigma n\mathbf{k}}^{\sigma_2 n' \mathbf{k}_2} - s_{\sigma_2\sigma}^e \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma n' \mathbf{k}}) + M_n^{n'} (s_{\sigma\sigma_1}^e \bar{C}_{\sigma\mathbf{k}}^{\sigma_2\mathbf{k}_2} - s_{\sigma_2\sigma}^e \bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma\mathbf{k}})] d^3\mathbf{k} \right\}, \quad (16c)$$

$$\left(-i\hbar\frac{\partial}{\partial t} + E_{\mathbf{k}_2} - E_{\mathbf{k}_1}\right)\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2} = Q_{\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}}^{\text{III}} + Q_{\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}}^{\text{IV}} + Q_{\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}}^{\text{V}}, \quad (16d)$$

where

$$Q_{\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}}^{\text{III}} = J_{sd} \sum_{n\sigma} \left\{ \frac{1}{(2\pi)^3} \int_{\text{BZ}} [\mathbf{S}_{nn_1} \cdot s_{\sigma\sigma_1}^e (\bar{K}_{\sigma n\mathbf{k}}^{\sigma_2 n_2 \mathbf{k}_2} + \bar{C}_{\sigma\mathbf{k}}^{\sigma_2\mathbf{k}_2} M_n^{n_2}) - \mathbf{S}_{n_2 n} \cdot s_{\sigma_2\sigma}^e (\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma n\mathbf{k}} + \bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma} M_{n_1}^n)] d^3\mathbf{k} \right. \\ \left. + (\mathbf{S}_{nn_1} \cdot s_{\sigma\sigma_1}^e C_{\sigma\mathbf{k}_2}^{\sigma_2} M_n^{n_2} - \mathbf{S}_{n_2 n} \cdot s_{\sigma_2\sigma}^e C_{\sigma_1\mathbf{k}_1}^{\sigma} M_{n_1}^n) \right\}, \quad (17a)$$

$$Q_{\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}}^{\text{IV}} = J_{sd} \sum_{nn'\sigma} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} [n_{Mn} (s_{\sigma\sigma_1}^e \bar{K}_{\sigma n\mathbf{k}}^{\sigma_2 n_2 \mathbf{k}_2} - s_{\sigma_2\sigma}^e \bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma n' \mathbf{k}}) - M_{n_1}^{n_2} (s_{\sigma\sigma_1}^e C_{\sigma\mathbf{k}_2}^{\sigma_2} - s_{\sigma_2\sigma}^e C_{\sigma_1\mathbf{k}_1}^{\sigma})] \right. \\ \left. - \frac{M_{n_1}^{n_2}}{(2\pi)^3} \int_{\text{BZ}} [(s_{\sigma\sigma_1}^e \bar{K}_{\sigma n\mathbf{k}}^{\sigma_2 n' \mathbf{k}_2} - s_{\sigma_2\sigma}^e \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma n' \mathbf{k}}) + M_n^{n'} (s_{\sigma\sigma_1}^e \bar{C}_{\sigma\mathbf{k}}^{\sigma_2\mathbf{k}_2} - s_{\sigma_2\sigma}^e \bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma\mathbf{k}})] d^3\mathbf{k} \right\}, \quad (17b)$$

$$Q_{\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}}^{\text{V}} = -J_{sd} \sum_{n\sigma\sigma'} s_{\sigma\sigma'}^e \cdot \left\{ \frac{C_{\sigma_1\mathbf{k}_1}^{\sigma'}}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{\sigma n\mathbf{k}}^{\sigma_2 n_2 \mathbf{k}_2} - \mathbf{S}_{n_2 n} \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma n\mathbf{k}}) d^3\mathbf{k} + C_{\sigma\mathbf{k}_2}^{\sigma_2} C_{\sigma_1\mathbf{k}_1}^{\sigma'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2 n} M_{n_1}^n) \right. \\ \left. + \frac{1}{(2\pi)^3} \int_{\text{BZ}} [C_{\sigma\mathbf{k}_2}^{\sigma_2} (\mathbf{S}_{nn_1} \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma' n_2 \mathbf{k}} - \mathbf{S}_{n_2 n} \bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma' n\mathbf{k}}) - C_{\sigma\mathbf{k}}^{\sigma'} (\mathbf{S}_{nn_1} \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2} - \mathbf{S}_{n_2 n} \bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n\mathbf{k}})] d^3\mathbf{k} \right. \\ \left. + \frac{1}{(2\pi)^3} \int_{\text{BZ}} (C_{\sigma_1\mathbf{k}_1}^{\sigma'} \bar{C}_{\sigma\mathbf{k}}^{\sigma_2\mathbf{k}_2} + C_{\sigma\mathbf{k}_2}^{\sigma_2} \bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma\mathbf{k}}) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2 n} M_{n_1}^n) d^3\mathbf{k} \right. \\ \left. + \frac{\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2}}{(2\pi)^6} \iint_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{\sigma n\mathbf{k}}^{\sigma' n_2 \mathbf{k}'} - \mathbf{S}_{n_2 n} \bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma' n\mathbf{k}'}) + \bar{C}_{\sigma\mathbf{k}}^{\sigma'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2 n} M_{n_1}^n)] d^3\mathbf{k}' d^3\mathbf{k} \right\}. \quad (17c)$$

As usual for large systems, we have treated the momenta \mathbf{k} in the above equations as continuous variables and sums over \mathbf{k} are represented as integrals according to the standard rule:

$$\sum_{\mathbf{k}} \rightarrow \frac{V}{(2\pi)^3} \int_{\text{BZ}} d^3\mathbf{k}, \quad (18)$$

where the integral extends over the first Brillouin zone. Note that the correlations $\delta\langle c_{\sigma_1\mathbf{k}_1}^\dagger c_{\sigma_2\mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle$ and $\delta\langle c_{\sigma_1\mathbf{k}_1}^\dagger c_{\sigma_2\mathbf{k}_2} e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle$ have been introduced in Eqs. (13b) and (13c) only for the case $\mathbf{k}_3 \neq 0$, which correspond to the off-diagonal elements with $\mathbf{k}_1 \neq \mathbf{k}_2$ of $\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}$ and $\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2}$. This restriction can be dropped when going over to integrals as the components of $\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2}$ and $\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}$ with $\mathbf{k}_1 = \mathbf{k}_2$ either couple only to themselves [cf. Eqs. (16c) and (16d)] or appear under integrals where they represent a set of measure zero and thus do not affect other dynamical variables.

It should be noted that the volume V does not appear explicitly in Eqs. (16), which implies that when the dynamical variables are initially $\mathcal{O}(1)$ in the volume V , this will hold true for all times. For the observables $M_{n_1}^{n_2}$ and $C_{\sigma_1\mathbf{k}_1}^{\sigma_2}$ that describe occupations or coherences, it thus follows directly from their definitions that they scale with the volume as $\mathcal{O}(1)$. In order to

determine initial values for $\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2}$ and $\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}$, a more detailed characterization of the initial state is required. For many typical DMS (paramagnetic as well as ferromagnetic samples), calculations based on the mean-field and virtual crystal (MF and VC) approximation are commonly used and known to yield reasonable results for ground-state properties.³⁰ On this level of description, $\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2}$ and $\bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2}$ are zero. Immediately after a short pulse excitation starting from the MF and VC ground state, the correlations did not have the time to build up and thus the above equations (16) can be used with nonequilibrium initial values for $M_{n_1}^{n_2}$ and $C_{\sigma_1\mathbf{k}_1}^{\sigma_2}$ that result from the excitation and $\bar{C}_{\sigma_1\mathbf{k}_1}^{\sigma_2\mathbf{k}_2} = \bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2} = 0$ as initial values. Of course, a full modeling of the excitation process requires the inclusion of the valence bands and the coupling to the external laser field. The corresponding rather lengthy extension of Eqs. (16) is straightforward along the same lines discussed so far. The resulting equations can be found in Appendix C.

Whenever approximations are made in order to solve many-particle problems, the question arises as to whether central physical properties are maintained by the approximation. In particular, conservation laws play an important role in physical processes such as, e.g., the conservation of quantities such as

- (i) the total spin $\Sigma^{\text{total}} := \langle \sum_I \hat{S}^I + \sum_i \hat{s}_i^e \rangle$,
- (ii) the number of electrons $N_e := \sum_{\sigma\mathbf{k}} C_{\sigma\mathbf{k}}^\sigma$, and
- (iii) the energy $\mathcal{E} := \langle H_0 + H_{sd} \rangle$.

All the above quantities can be expressed by our dynamical variables for which we have derived the approximate equations of motion (16). By evaluating the time derivative of Σ^{total} , N_e , and \mathcal{E} according to these equations, it is straightforward but lengthy to show that these quantities are conserved also by the dynamics defined by our truncation scheme as long as the system is not externally driven. The conservation of the total spin, the number of electrons, and the energy is an important consistency requirement that is perfectly fulfilled by our approach.

IV. SPIN TRANSFER AND MOMENTUM REDISTRIBUTION FOR PARAMAGNETS

The spatially homogeneous equations of motion can be applied to both paramagnetic and ferromagnetic DMS. Yet, the true correlations \bar{C} and \bar{K} influence the spin dynamics of the system most prominently when a mean-field description of the exchange interaction yields no dynamics at all. This is the case for paramagnetic DMS at zero magnetic field since here the mean magnetization of the Mn atoms is initially zero (cf. Sec. V A). In this section, we shall discuss general properties of the spin dynamics in such systems. To be specific, we study the onset of the spin transfer between electrons and Mn atoms in a spatially homogeneous system where initially the electrons are fully spin polarized and the Mn spins are randomly oriented.

A. Initial conditions

The assumption of randomly oriented Mn spins in the absence of an external magnetic field, which is characteristic for a paramagnet, translates into the following initial values of the Mn variables at the initial time $t = 0$:

$$M_{n_1}^{n_2} |_{t=0} = \frac{1}{6} \delta_{n_1 n_2}. \quad (19a)$$

Assuming that initially the bottom of the conduction band is occupied up to a maximal energy E_m , a spin-polarized electron density corresponds to the initial condition

$$C_{\sigma_1 \mathbf{k}_1}^{\sigma_2} |_{t=0} = \delta_{\sigma_1 \sigma_2} \delta_{\sigma_1 \uparrow} \Theta(E_m - E_{\mathbf{k}_1}). \quad (19b)$$

As discussed before for typical DMS, it can be expected to be a good approximation to assume that immediately after a short pulse excitation from the ground state, the correlations \bar{C} and \bar{K} are zero, which translates to

$$\bar{C}_{\sigma_1 \mathbf{k}_1}^{\sigma_2 \mathbf{k}_2} |_{t=0} = \bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2} |_{t=0} = 0 \quad (19c)$$

as initial condition for Eqs. (16).

B. Onset of the dynamics

Starting from these initial conditions, the spin-polarized electrons interact with the Mn atoms and a spin transfer between these subsystems sets in. In order to analyze the onset of the dynamics in detail, it is instructive to expand all pertinent variables in powers of the time t up to $\mathcal{O}(t^2)$. Inserting these expansions into Eqs. (16) yields analytical expressions for M , C , \bar{C} , and \bar{K} , which allow us to evaluate the relative importance

and the mutual dependencies of these variables in the very early phase of dynamics.

In zeroth order $\mathcal{O}(t^0)$, M , C , \bar{C} , and \bar{K} are given by the initial conditions (19), respectively. Up to linear order in t , all variables remain constant with the exception of the electron Mn spin correlations \bar{K} , which evolve according to

$$\begin{aligned} \bar{K}_{\sigma_1 n_1 \mathbf{k}_1}^{\sigma_2 n_2 \mathbf{k}_2} = & t \frac{i J_{sd}}{12\hbar} [\delta_{n_1 n_2} \delta_{\sigma_1 \sigma_2} \delta_{\sigma_1 \uparrow} S_{n_1 n_1}^z (\Theta(E_m - E_{\mathbf{k}_2}) \\ & - \Theta(E_m - E_{\mathbf{k}_1})) + S_{n_1(n_1-1)}^+ \delta_{\sigma_2 \uparrow} \delta_{\sigma_1 \downarrow} \\ & \delta_{n_1(n_2+1)} \Theta(E_m - E_{\mathbf{k}_2}) \\ & - S_{(n_1-1)n_1}^+ \delta_{\sigma_2 \downarrow} \delta_{\sigma_1 \uparrow} \delta_{n_1(n_2-1)} \Theta(E_m - E_{\mathbf{k}_1})], \end{aligned} \quad (20)$$

where $S_{n_1 n_2}^+ = S_{n_1 n_2}^x + i S_{n_1 n_2}^y$ denote the elements of the raising matrix. The first-order dynamics arise only from the first term on the right-hand side of $Q_{\bar{K}}^{\text{III}}$ in Eqs. (17a), namely,

$$J_{sd} \sum_{n\sigma} (S_{nn_1} \cdot s_{\sigma\sigma_1}^e C_{\sigma\mathbf{k}_2}^{\sigma_2} M_n^{n_2} - S_{n_2 n} \cdot s_{\sigma_2\sigma}^e C_{\sigma_1 \mathbf{k}_1}^{\sigma} M_{n_1}^n). \quad (21)$$

This term is in the disorder-averaged case the analog of Eq. (8a) and, like in Eq. (8a), reflects the exact reduction of the one-Mn-site density matrices to density matrices with only a single Mn operator. In the order $\mathcal{O}(t^2)$, only the terms proportional to t , i.e., the correlations \bar{K} , act as sources in the equations of motion. If these correlations were neglected, all other variables would be constant up to any order. Hence, under the special conditions studied here, the electron Mn spin correlations \bar{K} play a pivotal role for the onset of the spin transfer.

The change of the Mn and electron variables mediated by the correlations \bar{K} up to the second order in time is given by

$$\delta M_{n_1}^{n_2} = t^2 \delta_{n_1 n_2} \frac{p}{24\hbar^2} \frac{J_{sd}^2}{V_c^2} [(S_{n_1(n_1-1)}^+)^2 - (S_{(n_1+1)n_1}^+)^2], \quad (22a)$$

$$\begin{aligned} \delta C_{\sigma_1 \mathbf{k}_1}^{\sigma_2} = & t^2 \delta_{\sigma_1 \sigma_2} n_{\text{Mn}} \frac{35}{24\hbar^2} \frac{J_{sd}^2}{V_c} \\ & \times \left[\delta_{\sigma_1 \uparrow} \left(\frac{p}{2} - \frac{3}{2} \Theta(E_m - E_{\mathbf{k}_1}) \right) + \delta_{\sigma_1 \downarrow} p \right], \end{aligned} \quad (22b)$$

respectively, where V_c denotes the volume of a primitive lattice cell and

$$p := \frac{\int_{\text{BZ}} \Theta(E_m - E_{\mathbf{k}}) d^3 \mathbf{k}}{\int_{\text{BZ}} d^3 \mathbf{k}} \leq 1$$

is the fraction of the Brillouin zone that is initially occupied. Only the diagonal variables change in time, i.e., the onset of the spin transfer is not accompanied by a buildup of spin coherences such as, e.g., $C_{\downarrow \mathbf{k}}^\uparrow$ or $M_{5/2}^{3/2}$. It is also seen from Eqs. (20) and (22) that the sign of the exchange coupling J_{sd} determines the sign of the correlations \bar{K} but does not enter the values of the electronic or Mn occupations. In Eq. (22b), we can distinguish three terms: a term $\sim \Theta(E_m - E_{\mathbf{k}_1})$ which describes the depopulation of the initially occupied states and two terms which represent an inflow that depends on the spin of the target state but is independent of its \mathbf{k} vector. This implies that in the course of time all parts of the Brillouin zone will acquire finite occupations. Such a redistribution in \mathbf{k} space has to be expected for scattering processes caused by

a localized interaction such as, e.g., the exchange interaction with localized Mn spins given by the interaction Hamiltonian Eq. (3) because such a Hamiltonian is not translational invariant and, thus, does not preserve momentum. The interesting point here is that the transfer of occupation between states with different momenta is still present in our on average spatially homogeneous equations. It should be noted that, in contrast to the loss term, both gain terms are proportional to p . For carriers generated by near band-gap excitations with excess energies E_m between 10–100 meV the factor p is very small in the range of 10^{-5} – 10^{-3} . Consequently, for such conditions, an initially occupied state is rapidly depopulated, while the populations of initially unoccupied states takes place on a much longer time scale. Moreover, the two gain terms have different prefactors: at a given time, twice as many spin-up than spin-down occupations are built up.

The initial dynamics up to second order in time for the initial conditions (19) are illustrated in Fig. 1 for the case of a parabolic band $E_{\mathbf{k}} = \frac{\hbar^2 \mathbf{k}^2}{2m}$ with $m = 0.2m_e$, where m_e is the free-electron mass. Furthermore, we used $J_{sd} = 11$ meV nm³, $n_{\text{Mn}} = 1.7 \times 10^{-1}$ nm⁻³, $V_c = 5.7 \times 10^{-2}$ nm³, and $E_m = 96$ meV corresponding to $p = 3.4 \times 10^{-4}$. The material-specific parameters are typical values for Zn_{1-x}Mn_xTe,³¹ where the chosen Mn density n_{Mn} corresponds to $x = 0.01$. The expansion up to second order in time is, of course, expected to be quantitatively correct only for a few time steps where the absolute values of all quantities are not much changed. We found it, however, instructive to analyze the relative magnitudes of the changes of some relevant quantities in this initial phase of the dynamics. To this end, we plotted these quantities over some short but more or less arbitrary time. As all quantities in this second-order expansion evolve on parabolas, the relative magnitudes of the corresponding changes can be evaluated even when the range over that the plot is extended might be beyond the limit of validity of the expansion. Figure 1(a) displays the fraction of electrons with either spin up or down as well as the fraction of electrons with energies below or above E_m , i.e., in states with \mathbf{k} vectors that are initially occupied or unoccupied. It is seen that spin-up electrons are converted into spin-down electrons and that a redistribution from lower to higher kinetic energies takes place. It should be noted that the curvature of the parabolas that describe the spin conversion is proportional to n_{Mn} but independent of p . In contrast, the curvature of the parabolas representing the redistribution of kinetic energies depends on both n_{Mn} and p , and for our parameters it is stronger, implying that the momentum redistribution is initially faster than the spin flipping. Plotted in Fig. 1(b) is the corresponding change of the occupations of the Mn spin states. All Mn spin changes are up to second order in time proportional to p and independent of n_{Mn} . Consequently, for the low- p values considered here, the changes in the Mn occupations are rather low. The occupations of states with positive z component of the Mn spin rise, while states with negative z component are depopulated. The increase or decrease is stronger the larger the modulus of the z component. As expected for initially spin-up polarized electrons and unpolarized Mn spins, the exchange interaction leads to a net flow of spin from the electronic to the Mn subsystem. This is further illustrated in Fig. 1(c), where

the z components of the total electron spin $\Sigma^e := \sum_i \langle \hat{S}_i^z \rangle$ and Mn spin $\Sigma^{\text{Mn}} := \sum_I \langle \hat{S}_I^z \rangle$ normalized to the z component of the total spin are plotted. Finally, Fig. 1(d) displays separately different contributions to the energy, namely, the total kinetic energy of the electrons $\langle H_0 \rangle$, the total energy $\langle H_0 + H_{sd} \rangle$, the MF and VC exchange energy

$$\langle H_{sd} \rangle_{\text{VC}}^{\text{MF}} := J_{sd} n_{\text{Mn}} \sum_{nn'} \sum_{\sigma\sigma'} \mathbf{S}_{nn'} \cdot \mathbf{s}_{\sigma\sigma'}^e C_{\sigma\mathbf{k}}^{\sigma'} M_n^{\sigma'} \quad (23)$$

as well as the correlated part of the exchange energy $\langle H_{sd} \rangle_C := \langle H_{sd} \rangle - \langle H_{sd} \rangle_{\text{VC}}^{\text{MF}}$. Clearly, the changes of the kinetic energy and the correlation energy are much stronger than the change of the MF and VC energy [cf. also the inset of Fig. 1(d)]. This is related to the small value of p . For the other extreme of an initially fully occupied spin-up band, i.e., $p = 1$, the kinetic energy would be constant and the change in the correlation energy would exactly compensate the change of the MF and VC exchange energy. It is worth mentioning that the sign of the change of the MF and VC exchange energy is given by the sign of J_{sd} . This follows from Eq. (23) by noting that the carrier and Mn densities are up to second order in time, independent of the sign of J_{sd} . In all cases, the total energy is strictly conserved for all times.

C. Nonzero variables

As already hinted by the starting dynamics, the number of nonzero variables can be further reduced for the initial conditions discussed here. This can be seen by noting that within the equations of motion (16a)–(16d), the following variables form a closed subset if all other variables are initially zero:

$$M_n^{\sigma}, \quad C_{\sigma\mathbf{k}}^{\sigma}, \quad \bar{C}_{\sigma\mathbf{k}_1}^{\sigma\mathbf{k}_2}, \quad \bar{K}_{\uparrow(n-1)\mathbf{k}_1}^{\downarrow n\mathbf{k}_2}, \quad \bar{K}_{\downarrow(n+1)\mathbf{k}_1}^{\uparrow n\mathbf{k}_2}, \quad \bar{K}_{\sigma n\mathbf{k}_1}^{\sigma n\mathbf{k}_2}, \quad (24)$$

which implies that all other variables remain zero when they are initially zero. This is the case for the initial conditions given by Eqs. (19). We thus only need to consider the variables in Eq. (24) to describe the dynamics that start from these conditions.

Limiting the set of dynamical variables according to Eq. (24) simplifies the equations of motion (16a) and (16b) for the Mn and electron variables to

$$-i\hbar \frac{\partial}{\partial t} M_n^{\sigma} = \delta_{n_1 n_2} \frac{J_{sd}}{(2\pi)^6} \sum_{\sigma\sigma'} \mathbf{s}_{\sigma\sigma'}^e \cdot \int \int_{\text{BZ}} d^3\mathbf{k}' d^3\mathbf{k} \times (\mathbf{S}_{n n_1} \bar{K}_{\sigma n\mathbf{k}}^{\sigma' n_1 \mathbf{k}'} - \mathbf{S}_{n_1 n} \bar{K}_{\sigma n_1 \mathbf{k}}^{\sigma' n \mathbf{k}'}), \quad (25a)$$

$$-i\hbar \frac{\partial}{\partial t} C_{\sigma_1 \mathbf{k}_1}^{\sigma_2} = \delta_{\sigma_1 \sigma_2} \frac{J_{sd}}{(2\pi)^3} n_{\text{Mn}} \sum_{nn'} \mathbf{S}_{nn'} \cdot \int_{\text{BZ}} d^3\mathbf{k} \times \left[\mathbf{s}_{\sigma\sigma_1}^e (\bar{K}_{\sigma n\mathbf{k}}^{\sigma' n' \mathbf{k}_1} + M_n^{\sigma'} \bar{C}_{\sigma\mathbf{k}}^{\sigma_1 \mathbf{k}_1}) - \mathbf{s}_{\sigma_1 \sigma}^e (\bar{K}_{\sigma_1 n \mathbf{k}_1}^{\sigma' n' \mathbf{k}} + M_n^{\sigma'} \bar{C}_{\sigma_1 \mathbf{k}_1}^{\sigma \mathbf{k}}) \right]. \quad (25b)$$

Interestingly, all terms of the form C times M , i.e., MF and VC contributions, cancel out although the expectation value of the Mn spin acquires nonzero values in the course of time. As we have already seen perturbatively up to the order $\mathcal{O}(t^2)$, coherences between different electron or Mn spin states do

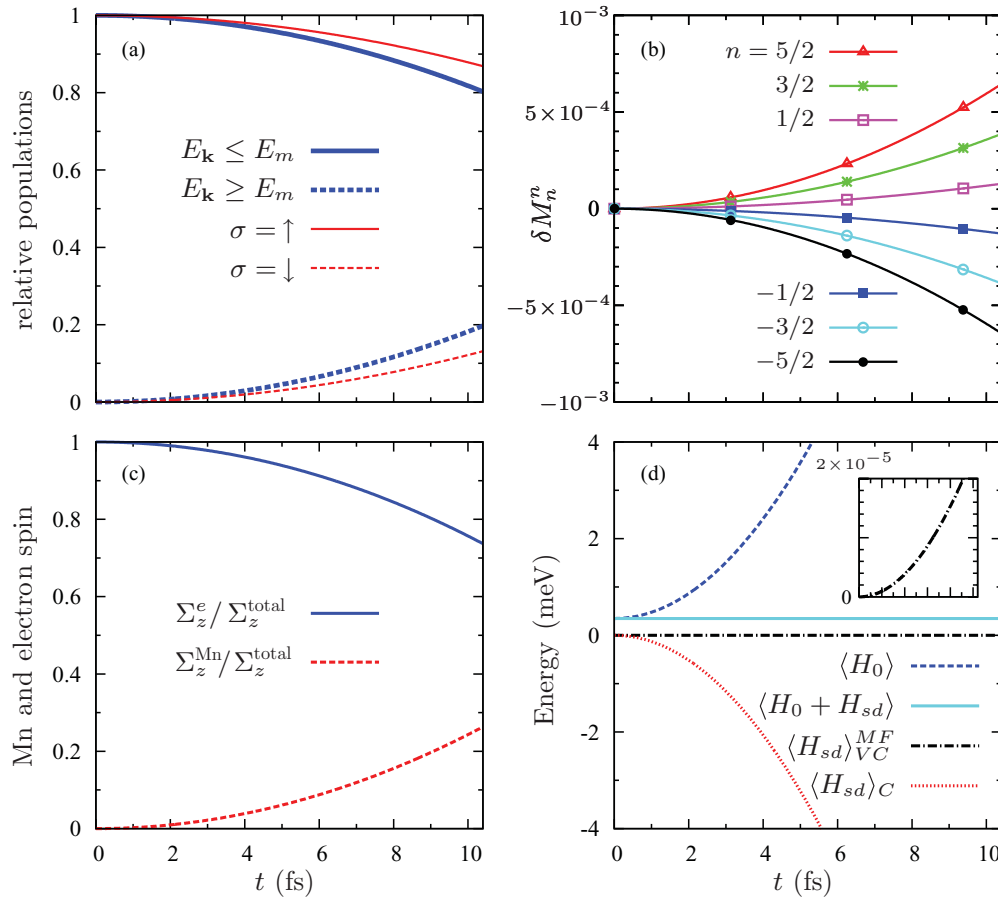


FIG. 1. (Color online) Time evolution up to second order in time of (a) fraction of spin-up or spin-down electrons and fraction of electrons with kinetic energies below or above E_m (as indicated); (b) change of the occupation of Mn spin states; (c) z component of the total electron (Σ_z^e) and Mn (Σ_z^{Mn}) spin normalized to the z component of the total spin (Σ_z^{total}); (d) total kinetic energy of the electrons $\langle H_0 \rangle$, total energy $\langle H_0 + H_{sd} \rangle$, MF and VC exchange energy $\langle H_{sd} \rangle_{VC}^{MF}$, correlated part of the exchange energy $\langle H_{sd} \rangle_C$. The inset in (d) shows the time evolution of $\langle H_{sd} \rangle_{VC}^{MF}$ with an adjusted energy scale.

not arise. The spin transfer between electrons and Mn atoms is mediated by the correlations \bar{K} and \bar{C} only.

Note that for different initial conditions, e.g., for a ferromagnetic system, nonzero coherences may build up, the equations of motion of M and C may contain source terms of the form C times M . Moreover, the reduction of variables to the set in Eqs. (24) is not applicable for other initial conditions.

V. LIMITING CASES

Spin dynamics in ferromagnetic DMS are often treated within the mean-field and virtual approximation. Although these are different approximations, usually they are applied together.¹² The spatially homogeneous equations of motion (16) contain the MF and VC dynamics as a limiting case. In the following, we derive this limit and also compare our equations with the results of theories that invoke only one of these two approximations. We show that both approximations fail to describe a spin transfer between carriers and Mn atoms in homogeneous systems when initially the Mn magnetization is zero.

A. Mean-field and virtual crystal approximation

Within the virtual crystal approximation, the localized character of the Mn atoms is neglected.¹² The electrons interact with Mn spins, which are homogeneously smeared out over the whole crystal. Then, the exchange Hamiltonian is spatially homogeneous and reads as

$$H_{sd}^{\text{VC}} = \frac{J_{sd}}{V} \sum_{Ii} \hat{\mathbf{S}}_I \cdot \hat{\mathbf{s}}_i^e. \quad (26)$$

Formally, the virtual crystal Hamiltonian (26) is obtained from the exchange Hamiltonian (3) by replacing the factors $\delta(\mathbf{r}_i - \mathbf{R}_I)$ by $1/V$. In Eq. (3), these factors contain the information on the Mn atom positions. Applying additionally the mean-field approximation implies that the Mn spins are only influenced by the expectation values of the electron spins and vice versa.¹² Thus, Eq. (26) transforms to the following MF and VC exchange Hamiltonian:

$$H_{sd}^{\text{MFVC}} = \frac{J_{sd}}{V} \sum_{Ii} (\hat{\mathbf{S}}_I \cdot \langle \hat{\mathbf{s}}_i^e \rangle + \langle \hat{\mathbf{S}}_I \rangle \cdot \hat{\mathbf{s}}_i^e - \langle \hat{\mathbf{S}}_I \rangle \cdot \langle \hat{\mathbf{s}}_i^e \rangle). \quad (27)$$

Starting with this Hamiltonian leads directly to a closed set of equations of motion. As no Mn position is distinguished in the Hamiltonian (27), all expectation values calculated from these equations do not depend on these positions. Thus, averaging over the random distribution of Mn atoms has no effect on the resulting equations that resemble our equations of motion (16) for the spatially homogeneous case except for one important change: the true correlations \bar{C} and \bar{K} are absent. Thus, we only need to consider the Mn variables and the electronic density matrices. Their equations of motion in the MF and VC approximation read as

$$-i\hbar \frac{\partial}{\partial t} M_{n_1}^{n_2} = J_{sd} \frac{\Sigma^e}{V} \cdot \sum_n (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n), \quad (28a)$$

$$-i\hbar \frac{\partial}{\partial t} C_{\sigma_1 \mathbf{k}_1}^{\sigma_2} = J_{sd} \frac{\Sigma^{\text{Mn}}}{V} \cdot \sum_{\sigma} (\mathbf{s}_{\sigma\sigma_1}^e C_{\sigma \mathbf{k}_1}^{\sigma_2} - \mathbf{s}_{\sigma_2\sigma}^e C_{\sigma_1 \mathbf{k}_1}^{\sigma}), \quad (28b)$$

where Σ^e/V and Σ^{Mn}/V are the electron and Mn spin densities, respectively. The former equation of motion is equivalent to an undamped *Landau-Lifshitz-Gilbert* equation,³² which is commonly used to describe spin precession in ferromagnets. The spin-transfer mechanism discussed in Sec. IV, however, can not be described within the MF and VC approximation since the correlations \bar{K} and \bar{C} , which are the only sources for the spin transfer in this case, vanish [cf. Eqs. (25)].

B. Mean-field approximation

When only the mean-field approximation is applied without virtual crystal approximation, then we have to replace H_{sd} in Eq. (3) by its mean-field counterpart. Let us first concentrate on the case of given Mn positions without disorder averaging. Then, the resulting Hamiltonian reads as

$$H_{sd}^{\text{MF}} = J_{sd} \sum_i (\hat{\mathbf{S}}_I \cdot \langle \hat{\mathbf{s}}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I) \rangle + \langle \hat{\mathbf{S}}_I \rangle \cdot \hat{\mathbf{s}}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I) - \langle \hat{\mathbf{S}}_I \rangle \cdot \langle \hat{\mathbf{s}}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I) \rangle), \quad (29)$$

where the expectation values involve only the quantum mechanical average for fixed Mn positions. The Heisenberg equations derived from this Hamiltonian provide a closed set for $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle$ and $\langle \hat{P}_{n_1 n_2}^I \rangle$, which can be obtained from Eqs. (7a) and (7b) by the replacement

$$\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I \rangle \longrightarrow \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \langle \hat{P}_{n_1 n_2}^I \rangle.$$

As can be easily seen, all source terms in these equations vanish when the dynamical variables are either of the form

$$\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle = \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle \delta_{\sigma_1 \sigma_2}, \quad \langle \hat{P}_{n_1 n_2}^I \rangle = \langle \hat{P}_{n_1 n_1}^I \rangle \delta_{n_1 n_2} \quad (30)$$

or when

$$\langle \hat{P}_{n_1 n_2}^I \rangle = \frac{1}{6} \delta_{n_1 n_2} \quad \text{without restrictions to } \langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} \rangle. \quad (31)$$

When either of these relations holds initially, as is the case, e.g., for a paramagnet where Eq. (31) applies, then this will be true for all times because these variables will not change at all. Thus, the mean-field theory is unable to describe the spin transfer in a paramagnet with initially spin-polarized carriers and zero initial Mn magnetization for any given configuration of Mn atoms. Averaging this vanishing spin transfer over a

random distribution of Mn positions obviously can not lead to a finite spin flow in an on average homogeneous system.

C. Virtual crystal approximation

The above analysis revealed that correlations beyond the mean-field level are crucial for the spin transfer when the initial Mn magnetization is zero. A model based on the virtual crystal Hamiltonian H_{sd}^{VC} describes such correlations when the mean-field approximation is not applied additionally. It is tempting to think of the virtual crystal Hamiltonian H_{sd}^{VC} in Eq. (26) as a spatially averaged version of the exchange Hamiltonian H_{sd} in Eq. (3) that generates the dynamics in an on average spatially homogeneous system. However, the dynamics obtained from H_{sd}^{VC} significantly deviates from our results [Eqs. (16)] that were derived from the full Hamiltonian H_{sd} for an on average spatially homogeneous system. This can be made explicit by starting with H_{sd}^{VC} instead of H_{sd} and repeating all the steps that led to Eqs. (16). As discussed already for the MF and VC dynamics in the virtual crystal model, all expectation values are independent of the positions of the Mn atoms and therefore the disorder averaging is redundant in this model. Before going over to the limit $V \rightarrow \infty$, the resulting equation of motion for the Mn variables $M_{n_1}^{n_2}$ reads as

$$-i\hbar \frac{\partial}{\partial t} M_{n_1}^{n_2} = \frac{J_{sd}}{(2\pi)^3} \sum_{n\sigma\sigma'} \int_{\text{BZ}} \left[C_{\sigma \mathbf{k}}^{\sigma'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) + \frac{1}{V} (\mathbf{S}_{nn_1} K_{\sigma n \mathbf{k}}^{\sigma' n_2} - \mathbf{S}_{n_2n} K_{\sigma n_1 \mathbf{k}}^{\sigma' n}) \right] \cdot \mathbf{s}_{\sigma\sigma'}^e d^3 \mathbf{k}, \quad (32)$$

where the correlation $K_{\sigma_1 n_1 \mathbf{k}}^{\sigma_2 n_2}$ has been defined in Eq. (15d). By comparing this result with our Eq. (16a), we note that the correlations \bar{C} and \bar{K} are missing in Eq. (32), while all contributions involving only C and M remain unchanged. The term on the right-hand side of Eq. (32) containing $K_{\sigma_1 n_1 \mathbf{k}}^{\sigma_2 n_2}$ would for finite V also appear in Eq. (16a) but has been dropped as it does not contribute in the limit $V \rightarrow \infty$ (see below). Analogous changes apply to the equation of motion (16b) for the electronic density matrix $C_{\sigma_1 \mathbf{k}_1}^{\sigma_2}$.

Instead of discussing these changes in general, we would like to focus here only on the resulting consequences for the initial spin-transfer dynamics that sets in starting from the initial conditions in Eqs. (19). In this case, it turns out that by using the virtual crystal approximation, the driving term for the correlations K to first order in t coincides with the source for \bar{K} in Eq. (17) evaluated at $\mathbf{k}_1 = \mathbf{k}_2$. Consequently, to linear order in t , the correlations $K_{\sigma_1 n_1 \mathbf{k}}^{\sigma_2 n_2}$ are given by the \mathbf{k} -diagonal parts of Eq. (20). By inserting these correlations into the equations of motion in the virtual crystal approximation for $M_{n_1}^{n_2}$ and $C_{\sigma_1 \mathbf{k}_1}^{\sigma_2}$, we obtain instead of Eqs. (22)

$$\delta M_{n_1}^{n_2} = t^2 \delta_{n_1 n_2} \frac{p}{24\hbar^2} \frac{J_{sd}^2}{V_c V} [(S_{n_1(n_1-1)}^+)^2 - (S_{(n_1+1)n_1}^+)^2], \quad (33a)$$

$$\delta C_{\sigma_1 \mathbf{k}_1}^{\sigma_2} = t^2 \delta_{\sigma_1 \sigma_2} n_{\text{Mn}} \frac{35}{24\hbar^2} \frac{J_{sd}^2}{V} \Theta(E_m - E_{\mathbf{k}_1}) (\delta_{\sigma_1 \downarrow} - \delta_{\sigma_1 \uparrow}). \quad (33b)$$

According to these equations, in the virtual crystal approximation the changes of $M_{n_1}^{n_2}$ and $C_{\sigma_1 \mathbf{k}_1}^{\sigma_2}$ scale as $\sim 1/V$. Thus, in the macroscopic limit $V \rightarrow \infty$, the virtual crystal approximation is unable to describe a spin transfer from initially spin-polarized carriers to the initially unpolarized Mn system although the correlations \bar{K} are nonzero. Obviously, the correlations \bar{K} that are off diagonal with respect to \mathbf{k} are decisive for the spin transfer. Therefore, for finite volumes V where also in the virtual crystal approximation a finite transfer is obtained, such results should be expected to be quantitatively doubtful as they have a wrong V scaling. Furthermore, also in contrast to Eqs. (22) in the virtual crystal approximation, there is no redistribution in momentum space. This is a consequence of the fact that the virtual crystal Hamiltonian is translational invariant. From this analysis, it is clearly seen that it makes a significant difference whether the dynamics in a spatially averaged Hamiltonian are considered or the dynamical equations of a spatially inhomogeneous system are averaged to describe an on average spatially homogeneous system.

VI. CONCLUSION

We have derived quantum kinetic equations of motion for the dynamics of diluted magnetic semiconductors. Our theory accounts for the exchange interaction between carriers and localized magnetic dopants beyond the mean-field and virtual crystal approximations. The main achievement is a systematic truncation scheme that exactly reduces density matrices involving two Mn spin operators for the same site to expectation values containing only a single Mn operator. Fundamental conservation laws, e.g., for total spin, particle numbers, and energy, are preserved when the hierarchy of equations of motion is truncated according to our scheme. Explicitly worked out are the cases of an on average spatially homogeneous single-band bulk system [Eqs. (16)], a spatially inhomogeneous single-band bulk system [Eqs. (7)], as well as the case of a spatially homogeneous multiband bulk system with laser coupling [Eqs. (C8)]. Also explained are the modifications needed to describe quantum wells or wires (cf. Appendix B).

For the spatially homogeneous single-band bulk case, we have analyzed how the spin transfer between initially spin-polarized carriers and unpolarized Mn atoms sets in. It turns out that the correct treatment of the density matrices with two Mn spin operators for the same site is decisive for the dynamics of genuine Mn-carrier correlations \bar{K} that provide the only driving term for the onset of this spin transfer. In a mean-field theory, this spin transfer can not be described as such Mn carrier correlations are disregarded. Even though the virtual crystal approximation yields finite correlations when it is not combined with the mean-field approximation, it turns out that the spin transfer predicted on this level of theory would vanish in the limit of an infinite crystal volume. Clearly, this prediction is unphysical in contrast to the results of our quantum kinetic equations. The decisive difference to our approach is that in the virtual crystal approximation, the Mn carrier correlations \bar{K} , which are off diagonal with respect to the involved momenta, are missing.

The spin transfer is in the quantum kinetic equations accompanied by a redistribution of the carriers in momentum space.

This reflects the fact that the exchange interaction is spatially localized and thus momentum is not conserved. Interestingly, this feature remains after the dynamical equations have been averaged over spatially homogeneous distributions of Mn atoms, which makes the dynamics spatially homogeneous on average. In contrast, the virtual crystal approximation, which starts from a translationally invariant model Hamiltonian, misses this momentum redistribution completely.

Our quantum kinetic equations of motion provide a widely applicable microscopic basis for future research on the dynamics of both paramagnetic and ferromagnetic DMS, which shall allow us also to address issues related to coherent phenomena that occur on short time scales.

APPENDIX A: DECOMPOSITION INTO CORRELATION FUNCTIONS

In this Appendix, we shall give for all quantities that are kept in our theory the explicit decomposition into factorized and irreducible parts, which form the basis for the truncation scheme that is described in Sec. III A3.

The decomposition for the two-Mn-site density matrices $\langle c_{\sigma_1 \mathbf{k}_1}^\dagger c_{\sigma_2 \mathbf{k}_2} P_{n_1 n_2}^{I'} P_{n_3 n_4}^I \rangle$ with $I' \neq I$ reads as

$$\begin{aligned} \langle c_{j_1}^\dagger c_{j_2} P^{I'} P^I \rangle &\stackrel{I \neq I'}{=} \langle c_{j_1}^\dagger c_{j_2} \rangle \langle P^{I'} \rangle \langle P^I \rangle + \langle P^I \rangle \delta \langle c_{j_1}^\dagger c_{j_2} P^{I'} \rangle \\ &+ \langle P^{I'} \rangle \delta \langle c_{j_1}^\dagger c_{j_2} P^I \rangle + \langle c_{j_1}^\dagger c_{j_2} \rangle \delta \langle P^{I'} P^I \rangle + \delta \langle c_{j_1}^\dagger c_{j_2} P^{I'} P^I \rangle, \end{aligned} \quad (\text{A1})$$

where we have combined the two indices σ and \mathbf{k} of the electronic operators c and c^\dagger to one index j and omitted the n indices of the Mn operators P^I for the sake of a shorter notation. Equation (A1) has to be regarded as the definition of the irreducible correlation function $\delta \langle c_{j_1}^\dagger c_{j_2} P^{I'} P^I \rangle$ as all other quantities that appear in that equation have been defined independently in the main part of the text. The five-point density matrices $\langle c_{j_1}^\dagger c_{j_2}^\dagger c_{j_3} c_{j_4} P^I \rangle$ are decomposed according to the following scheme:

$$\begin{aligned} \langle c_{j_1}^\dagger c_{j_2}^\dagger c_{j_3} c_{j_4} P^I \rangle &= (\langle c_{j_1}^\dagger c_{j_4} \rangle \langle c_{j_2}^\dagger c_{j_3} \rangle - \langle c_{j_1}^\dagger c_{j_3} \rangle \langle c_{j_2}^\dagger c_{j_4} \rangle) \langle P^I \rangle \\ &+ \langle c_{j_1}^\dagger c_{j_4} \rangle \delta \langle c_{j_2}^\dagger c_{j_3} P^I \rangle + \langle c_{j_2}^\dagger c_{j_3} \rangle \delta \langle c_{j_1}^\dagger c_{j_4} P^I \rangle \\ &- \langle c_{j_1}^\dagger c_{j_3} \rangle \delta \langle c_{j_2}^\dagger c_{j_4} P^I \rangle - \langle c_{j_2}^\dagger c_{j_4} \rangle \delta \langle c_{j_1}^\dagger c_{j_3} P^I \rangle \\ &+ \langle P^I \rangle \delta \langle c_{j_1}^\dagger c_{j_2}^\dagger c_{j_3} c_{j_4} \rangle + \delta \langle c_{j_1}^\dagger c_{j_2}^\dagger c_{j_3} c_{j_4} P^I \rangle, \end{aligned} \quad (\text{A2})$$

which defines $\delta \langle c_{j_1}^\dagger c_{j_2}^\dagger c_{j_3} c_{j_4} P^I \rangle$.

APPENDIX B: QUANTUM WELLS AND WIRES

The equations of motion which have been developed in the main part of this paper can only describe bulk systems where the conduction-band electrons can move freely in all directions. Nevertheless, our theoretical approach can be adopted to quantum wells and wires as will be detailed in the following. In such structures, the conduction-band electrons can move freely in the plane of the well (along the wire), but are confined in the direction(s) perpendicular to the well (wire). In the position representation, the eigenfunctions of the

electronic part of the Hamiltonian are of the form

$$\Phi_{\mathbf{k}_{\parallel}j\sigma}(\mathbf{r}) = \frac{1}{\sqrt{A}} e^{i\mathbf{k}_{\parallel}\mathbf{r}_{\parallel}} \eta_j(\mathbf{r}_{\perp}) u_{\sigma}(\mathbf{r}), \quad (\text{B1})$$

where \mathbf{k}_{\parallel} and \mathbf{r}_{\parallel} are the components of \mathbf{k} and \mathbf{r} in the directions parallel to the well (wire), \mathbf{r}_{\perp} are the components in the confined directions, η_j the envelope functions in the confined directions, and u_{σ} the lattice periodic functions. A denotes the area of the well (length of the wire). In this basis, the matrix elements of the operator $\hat{S}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I)$ in the exchange Hamiltonian in Eq. (3) have the form

$$\langle \Phi_{\mathbf{k}_{\parallel}j\sigma} | \hat{S}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I) | \Phi_{\mathbf{k}'_{\parallel}j'\sigma'} \rangle = \frac{1}{A} e^{i(\mathbf{k}'_{\parallel} - \mathbf{k}_{\parallel})\mathbf{R}_{\parallel I}} \times \eta_j(\mathbf{R}_{\perp I})^* \eta_{j'}(\mathbf{R}_{\perp I}) \mathbf{s}_{\sigma\sigma'}^e. \quad (\text{B2})$$

In order to follow the scheme presented in Sec. III, we require that $\eta(\mathbf{R}_{\perp I})$ does not depend on the perpendicular components $\mathbf{R}_{\perp I}$ of the Mn position vector \mathbf{R}_I . This can be achieved by using the approximation

$$\eta(\mathbf{R}_{\perp I}) \approx \frac{1}{\sqrt{D}}, \quad (\text{B3})$$

where D is the thickness of the well (the cross-sectional area of the wire), i.e., with respect to the spatial distribution of the Mn atoms in the confined direction the model is replaced by its average over the components of the Mn positions in these directions. Inserting Eq. (B3) into (B1) yields

$$\langle \Phi_{\mathbf{k}_{\parallel}j\sigma} | \hat{S}_i^e \delta(\mathbf{r}_i - \mathbf{R}_I) | \Phi_{\mathbf{k}'_{\parallel}j'\sigma'} \rangle \approx \frac{1}{V} e^{i(\mathbf{k}'_{\parallel} - \mathbf{k}_{\parallel})\mathbf{R}_{\parallel I}} \mathbf{s}_{\sigma\sigma'}^e, \quad (\text{B4})$$

where $V = AD$ is the volume of the well (wire). Due to the approximation given in Eq. (B3), the above matrix elements have the same structure as their counterparts in bulk systems with the only difference lying in the dimension of the \mathbf{k} vector: in the case of quantum wells, \mathbf{k}_{\parallel} is two dimensional, while for quantum wires \mathbf{k}_{\parallel} has only one independent component. Hence, the theoretical approach presented in Sec. III can be analogously applied to well and wire structures.

APPENDIX C: LASER-DRIVEN MULTIBAND DYNAMICS

In this section, we present an extended version of the spatially homogeneous equations of motion (16). The extended set additionally accounts for p -like valence bands, the dipole interaction between the light field of a laser beam and the carriers, and the exchange interaction between p -like holes and localized Mn spins. It allows us to explicitly model the optical excitation of spin-polarized carriers and the spin transfer between holes and Mn spins.

For the derivation of the extended set of equations of motion, we use the following Hamiltonian as a starting point:

$$H' = H_0 + H_{em} + H_{sd} + H_{pd}, \quad (\text{C1})$$

and

$$H_0 = \sum_{l\mathbf{k}} E_{l\mathbf{k}} c_{l\mathbf{k}}^{\dagger} c_{l\mathbf{k}} + \sum_{v\mathbf{k}} E_{v\mathbf{k}} d_{v\mathbf{k}}^{\dagger} d_{v\mathbf{k}} \quad (\text{C2})$$

describes the band structure of the host semiconductor, where l denotes s -like conduction bands, v p -like valence bands. $E_{l\mathbf{k}}$

and $E_{v\mathbf{k}}$ are positive electron and hole energies, respectively. Here,

$$H_{em} = - \sum_{l'l\mathbf{k}} \mathbf{E} \cdot \mathbf{M}_{l'l\mathbf{k}}^e c_{l\mathbf{k}}^{\dagger} c_{l'\mathbf{k}} - \sum_{vv'\mathbf{k}} \mathbf{E} \cdot \mathbf{M}_{vv'\mathbf{k}}^h d_{v\mathbf{k}}^{\dagger} d_{v'\mathbf{k}} - \sum_{lv\mathbf{k}} (\mathbf{E} \cdot \mathbf{M}_{lv\mathbf{k}} c_{l\mathbf{k}}^{\dagger} d_{v-\mathbf{k}}^{\dagger} + \mathbf{E} \cdot \mathbf{M}_{v\mathbf{k}} d_{v-\mathbf{k}} c_{l\mathbf{k}}) \quad (\text{C3})$$

denotes the dipole interaction between a spatially homogeneous, time-dependent electric field \mathbf{E} of a laser beam and the carriers. The dipole moments \mathbf{M} are given by

$$\begin{aligned} \mathbf{M}_{v\mathbf{k}} &:= -e \langle v\mathbf{k} | \mathbf{r} | l\mathbf{k} \rangle = \mathbf{M}_{v\mathbf{k}}^*, \\ \mathbf{M}_{l'l\mathbf{k}}^e &:= -e \langle l\mathbf{k} | \mathbf{r} | l'\mathbf{k} \rangle, \\ \mathbf{M}_{vv'-\mathbf{k}}^h &:= +e \langle v'\mathbf{k} | \mathbf{r} | v\mathbf{k} \rangle, \end{aligned}$$

where $|l\mathbf{k}\rangle$ and $|v\mathbf{k}\rangle$ are Bloch functions, \mathbf{r} the position vector, and e the positive elemental charge. H_{sd} is given in Eqs. (3) and (6), respectively. For an arbitrary set of conduction bands, the spin index σ in Eq. (6) is replaced by the conduction-band index l , and $\mathbf{s}_{ll'}^e$ is, thus, not necessarily the vector of Pauli matrices but becomes

$$\mathbf{s}_{ll'}^e := V e^{i(\mathbf{k}-\mathbf{k}')\mathbf{R}_I} \langle l\mathbf{k} | \hat{S} \delta(\mathbf{r} - \mathbf{R}_I) | l'\mathbf{k}' \rangle,$$

where \hat{S} is the spin operator in the electron picture. By analogy, H_{pd} reads as

$$\begin{aligned} H_{pd} &= J_{pd} \sum_{Ii} \hat{S}_I \cdot \hat{S}_i^h \delta(\mathbf{r}_i - \mathbf{R}_I) \\ &= \frac{J_{pd}}{V} \sum_{lnn'} \sum_{vv'} \mathbf{S}_{nn'} \cdot \mathbf{s}_{vv'}^h e^{i(\mathbf{k}'-\mathbf{k})\mathbf{R}_I} d_{v\mathbf{k}}^{\dagger} d_{v'\mathbf{k}'} \hat{P}_{nn'}^I, \end{aligned} \quad (\text{C4})$$

where the hole spin matrix $\mathbf{s}_{vv'}^h$ is defined by

$$\mathbf{s}_{vv'}^h := -V e^{i(\mathbf{k}'-\mathbf{k})\mathbf{R}_I} \langle v'\mathbf{k}' | \hat{S} \delta(\mathbf{r} - \mathbf{R}_I) | v\mathbf{k} \rangle.$$

This definition of the matrix elements leads to high formal symmetry between electrons and holes in the Hamiltonian.

With the new system Hamiltonian H' , we follow the procedure detailed in Sec. III B in order to obtain the corresponding extended set of equations of motion for a spatially homogeneous system in the macroscopic limit $V \rightarrow \infty$: at first we set up Heisenberg equations of motions for operators

$$\hat{P}_{n_1 n_2}^I, \quad c_{l_1 \mathbf{k}_1}^{\dagger} c_{l_2 \mathbf{k}_2}, \quad d_{v_1 \mathbf{k}_1}^{\dagger} d_{v_2 \mathbf{k}_2}, \quad d_{v_1 \mathbf{k}_1} c_{l_2 \mathbf{k}_2}.$$

We average these equations over both the quantum mechanical statistical operator and the random distribution of the Mn positions. This leads to equations of motion for the basic disorder-averaged variables

$$\langle \hat{P}_{n_1 n_2}^I \rangle, \quad \langle c_{l_1 \mathbf{k}_1}^{\dagger} c_{l_2 \mathbf{k}_2} \rangle, \quad \langle d_{v_1 \mathbf{k}_1}^{\dagger} d_{v_2 \mathbf{k}_2} \rangle, \quad \langle d_{v_1 \mathbf{k}_1} c_{l_2 \mathbf{k}_2} \rangle, \quad (\text{C5})$$

which are coupled to disorder-averaged density matrices of higher order, namely,

$$\begin{aligned} &\langle c_{l_1 \mathbf{k}_1}^{\dagger} c_{l_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle, \quad \langle d_{v_1 \mathbf{k}_1}^{\dagger} d_{v_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle, \\ &\langle d_{v_1 \mathbf{k}_1} c_{l_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i\mathbf{k}_3 \mathbf{R}_I} \rangle. \end{aligned} \quad (\text{C6a})$$

In analogy to Eqs. (13), these density matrices are expressed by their decomposition into the basic irreducible functions

given in Eqs. (C5) and true correlation functions. Setting up equations of motion for these correlation functions leads to an infinite hierarchy of equations of motion. Along the lines of Sec. III B, this hierarchy is truncated by applying a correlation expansion to all higher-order density matrices and, for the case of an on average spatially homogeneous system in the macroscopic limit, only keeping four basic irreducible variables and six correlation functions. The former are given by

$$M_{n_1}^{n_2} := \langle \hat{P}_{n_1 n_2}^I \rangle, \quad (C7a)$$

$$C_{l_1 \mathbf{k}}^{l_2} := \langle c_{l_1 \mathbf{k}}^\dagger c_{l_2 \mathbf{k}} \rangle, \quad (C7b)$$

$$D_{v_1 \mathbf{k}}^{v_2} := \langle d_{v_1 \mathbf{k}}^\dagger d_{v_2 \mathbf{k}} \rangle, \quad (C7c)$$

$$Y_{v_1 \mathbf{k}}^{l_2} := \langle d_{v_1 \mathbf{k}} c_{l_2 - \mathbf{k}} \rangle, \quad (C7d)$$

and represent Mn variables, electron and hole density matrices, and electron-hole coherences. The true correlations are

defined as

$$\bar{C}_{l_1 \mathbf{k}_1}^{l_2 \mathbf{k}_2} := V \delta \langle c_{l_1 \mathbf{k}_1}^\dagger c_{l_2 \mathbf{k}_2} e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_l} \rangle, \quad (C7e)$$

$$\bar{D}_{v_1 \mathbf{k}_1}^{v_2 \mathbf{k}_2} := V \delta \langle d_{v_1 \mathbf{k}_1}^\dagger d_{v_2 \mathbf{k}_2} e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_l} \rangle, \quad (C7f)$$

$$\bar{Y}_{v_1 \mathbf{k}_1}^{l_2 \mathbf{k}_2} := V \delta \langle d_{v_1 \mathbf{k}_1} c_{l_2 \mathbf{k}_2} e^{i(\mathbf{k}_2 + \mathbf{k}_1) \mathbf{R}_l} \rangle, \quad (C7g)$$

$$\bar{K}_{C_{l_1 n_1 \mathbf{k}_1}}^{l_2 n_2 \mathbf{k}_2} := V \delta \langle c_{l_1 \mathbf{k}_1}^\dagger c_{l_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_l} \rangle, \quad (C7h)$$

$$\bar{K}_{D_{v_1 n_1 \mathbf{k}_1}}^{v_2 n_2 \mathbf{k}_2} := V \delta \langle d_{v_1 \mathbf{k}_1}^\dagger d_{v_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i(\mathbf{k}_2 - \mathbf{k}_1) \mathbf{R}_l} \rangle, \quad (C7i)$$

$$\bar{K}_Y^{l_2 n_2 \mathbf{k}_2} := V \delta \langle d_{v_1 \mathbf{k}_1} c_{l_2 \mathbf{k}_2} \hat{P}_{n_1 n_2}^I e^{i(\mathbf{k}_2 + \mathbf{k}_1) \mathbf{R}_l} \rangle, \quad (C7j)$$

in analogy to the single-band correlation functions \bar{C} and \bar{K} defined in Eq. (15). The resulting closed set of equations of motion for these dynamical variables that applies for an optically driven spatially homogeneous multiband bulk system is given by

(i) equations for the Mn variables M :

$$\begin{aligned} -i\hbar \frac{\partial}{\partial t} M_{n_1}^{n_2} &= \frac{J_{sd}}{(2\pi)^3} \int_{\text{BZ}} \sum_{n'l'} s_{ll'}^e \cdot \left[\left(C_{l\mathbf{k}}^{l'} + \int_{\text{BZ}} \frac{\bar{C}_{l\mathbf{k}}^{l'\mathbf{k}'}}{(2\pi)^3} d^3\mathbf{k}' \right) (\mathbf{S}_{nn_1} M_{n_1}^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^{n_2}) + \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{C_{l\mathbf{k}}}^{l'n_2\mathbf{k}'} - \mathbf{S}_{n_2n} \bar{K}_{C_{l\mathbf{k}}}^{l'n_2\mathbf{k}'}) \frac{d^3\mathbf{k}'}{(2\pi)^3} \right] d^3\mathbf{k} \\ &+ \frac{J_{pd}}{(2\pi)^3} \int_{\text{BZ}} \sum_{nv'} s_{vv'}^h \cdot \left[\left(D_{v\mathbf{k}}^{v'} + \int_{\text{BZ}} \frac{\bar{D}_{v\mathbf{k}}^{v'\mathbf{k}'}}{(2\pi)^3} d^3\mathbf{k}' \right) (\mathbf{S}_{nn_1} M_{n_1}^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^{n_2}) \right. \\ &\left. + \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{D_{v\mathbf{k}}}^{v'n_2\mathbf{k}'} - \mathbf{S}_{n_2n} \bar{K}_{D_{v\mathbf{k}}}^{v'n_2\mathbf{k}'}) \frac{d^3\mathbf{k}'}{(2\pi)^3} \right] d^3\mathbf{k}; \end{aligned} \quad (C8a)$$

(ii) equations for the electron two-point density matrices C :

$$\begin{aligned} \left(-i\hbar \frac{\partial}{\partial t} + E_{l_2 \mathbf{k}_1} - E_{l_1 \mathbf{k}_1} \right) C_{l_1 \mathbf{k}_1}^{l_2} &= \sum_v \mathbf{E} \cdot (\mathbf{M}_{v l_2 \mathbf{k}_1}^* (Y_{v - \mathbf{k}_1}^{l_1})^* - \mathbf{M}_{v l_1 \mathbf{k}_1} Y_{v - \mathbf{k}_1}^{l_2}) + \sum_l \mathbf{E} \cdot (\mathbf{M}_{l_2 l \mathbf{k}_1}^e C_{l_1 \mathbf{k}_1}^{l_2} - \mathbf{M}_{l l_1 \mathbf{k}_1}^e C_{l_1 \mathbf{k}_1}^{l_2}) \\ &+ J_{sd} n_{\text{Mn}} \sum_{nn'} \mathbf{S}_{nn'} \cdot \left\{ M_{n'}^{n'} \left[s_{ll_1}^e \left(C_{l\mathbf{k}_1}^{l_2} + \int_{\text{BZ}} \frac{\bar{C}_{l\mathbf{k}}^{l_2 \mathbf{k}_1}}{(2\pi)^3} d^3\mathbf{k} \right) - s_{l_2 l}^e \left(C_{l_1 \mathbf{k}_1}^{l_2} + \int_{\text{BZ}} \frac{\bar{C}_{l_1 \mathbf{k}_1}^{l_2 \mathbf{k}_1}}{(2\pi)^3} d^3\mathbf{k} \right) \right] \right. \\ &\left. + \int_{\text{BZ}} (s_{ll_1}^e \bar{K}_{C_{l\mathbf{k}}}^{l_2 n' \mathbf{k}_1} - s_{l_2 l}^e \bar{K}_{C_{l_1 \mathbf{k}_1}}^{l_2 n' \mathbf{k}_1}) \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}; \end{aligned} \quad (C8b)$$

(iii) equations for the hole two-point density matrices D :

$$\begin{aligned} \left(-i\hbar \frac{\partial}{\partial t} + E_{v_2 \mathbf{k}_1} - E_{v_1 \mathbf{k}_1} \right) D_{v_1 \mathbf{k}_1}^{v_2} &= \sum_l \mathbf{E} \cdot (\mathbf{M}_{v_2 l - \mathbf{k}_1}^* (Y_{v_1 \mathbf{k}_1}^l)^* - \mathbf{M}_{v_1 l - \mathbf{k}_1} Y_{v_2 \mathbf{k}_1}^l) + \sum_v \mathbf{E} \cdot (\mathbf{M}_{v_2 v \mathbf{k}_1}^h D_{v_1 \mathbf{k}_1}^{v_2} - \mathbf{M}_{v v_1 \mathbf{k}_1}^h D_{v_1 \mathbf{k}_1}^{v_2}) \\ &+ J_{pd} n_{\text{Mn}} \sum_{nn'} \mathbf{S}_{nn'} \cdot \left\{ M_{n'}^{n'} \left[s_{vv_1}^h \left(D_{v_1 \mathbf{k}_1}^{v_2} + \int_{\text{BZ}} \frac{\bar{D}_{v\mathbf{k}}^{v_2 \mathbf{k}_1}}{(2\pi)^3} d^3\mathbf{k} \right) - s_{v_2 v}^h \left(D_{v_1 \mathbf{k}_1}^{v_2} + \int_{\text{BZ}} \frac{\bar{D}_{v_1 \mathbf{k}_1}^{v_2 \mathbf{k}_1}}{(2\pi)^3} d^3\mathbf{k} \right) \right] \right. \\ &\left. + \int_{\text{BZ}} (s_{vv_1}^h \bar{K}_{D_{v\mathbf{k}}}^{v_2 n' \mathbf{k}_1} - s_{v_2 v}^h \bar{K}_{D_{v_1 \mathbf{k}_1}}^{v_2 n' \mathbf{k}_1}) \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}; \end{aligned} \quad (C8c)$$

(iv) equations for the electron-hole coherences Y :

$$\begin{aligned} \left(-i\hbar \frac{\partial}{\partial t} + E_{v_1 \mathbf{k}_1} + E_{l_2 - \mathbf{k}_1} \right) Y_{v_1 \mathbf{k}_1}^{l_2} \\ = \mathbf{E} \cdot \mathbf{M}_{l_2 v_1 - \mathbf{k}_1} + \sum_l \mathbf{E} \cdot (\mathbf{M}_{l_2 l - \mathbf{k}_1}^e Y_{v_1 \mathbf{k}_1}^l - \mathbf{M}_{l v_1 - \mathbf{k}_1} C_{l - \mathbf{k}_1}^{l_2}) + \sum_v \mathbf{E} \cdot (\mathbf{M}_{v_1 v \mathbf{k}_1}^h Y_{v_1 \mathbf{k}_1}^{l_2} - \mathbf{M}_{l_2 v - \mathbf{k}_1} D_{v \mathbf{k}_1}^{v_1}) \end{aligned}$$

$$\begin{aligned}
& -n_{\text{Mn}} \sum_{nn'} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} \left[J_{pd} \sum_v s_{v\mathbf{k}_1}^h \left(Y_{v\mathbf{k}_1}^{l_2} + \int_{\text{BZ}} \frac{\bar{Y}_{v\mathbf{k}}^{l_2-\mathbf{k}_1}}{(2\pi)^3} d^3\mathbf{k} \right) + J_{sd} \sum_l s_{l_2l}^e \left(Y_{v\mathbf{k}_1}^l + \int_{\text{BZ}} \frac{\bar{Y}_{v\mathbf{k}_1}^{lk}}{(2\pi)^3} d^3\mathbf{k} \right) \right] \right. \\
& \left. + \int_{\text{BZ}} \left(J_{pd} \sum_v s_{v\mathbf{k}_1}^h \bar{K}_{Y_{v\mathbf{k}_1}^{l_2n'-\mathbf{k}_1}} + J_{sd} \sum_l s_{l_2l}^e \bar{K}_{Y_{v\mathbf{k}_1}^{ln'\mathbf{k}_1}} \right) \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}; \quad (\text{C8d})
\end{aligned}$$

(v) equations for correlations \bar{C} involving electron operators and Mn position phase factors:

$$\begin{aligned}
\left(-i\hbar \frac{\partial}{\partial t} + E_{l_2\mathbf{k}_2} - E_{l_1\mathbf{k}_1} \right) \bar{C}_{l_1\mathbf{k}_1}^{l_2\mathbf{k}_2} &= \sum_v \mathbf{E} \cdot (\mathbf{M}_{v l_2\mathbf{k}_2}^* (\bar{Y}_{v-\mathbf{k}_2}^{l_1\mathbf{k}_1})^* - \mathbf{M}_{v l_1\mathbf{k}_1} \bar{Y}_{v-\mathbf{k}_1}^{l_2\mathbf{k}_2}) + \sum_l \mathbf{E} \cdot (\mathbf{M}_{l_2l\mathbf{k}_2}^e \bar{C}_{l_1\mathbf{k}_1}^{l_2\mathbf{k}_2} - \mathbf{M}_{l_1\mathbf{k}_1}^e \bar{C}_{l_1\mathbf{k}_1}^{l_2\mathbf{k}_2}) \\
&+ J_{sd} \sum_{nn'l} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} [s_{l_1l}^e (n_{\text{Mn}} \bar{C}_{l_1\mathbf{k}_1}^{l_2\mathbf{k}_2} + C_{l_2\mathbf{k}_2}^{l_2}) - s_{l_2l}^e (n_{\text{Mn}} \bar{C}_{l_1\mathbf{k}_1}^{l_2\mathbf{k}_2} + C_{l_1\mathbf{k}_1}^l)] \right. \\
&\left. + \int_{\text{BZ}} [(s_{l_1l}^e \bar{K}_{C_{l_1n\mathbf{k}}}^{l_2n'\mathbf{k}_2} - s_{l_2l}^e \bar{K}_{C_{l_1n\mathbf{k}_1}}^{ln'\mathbf{k}_1}) + M_n^{n'} (s_{l_1l}^e \bar{C}_{l_1\mathbf{k}_1}^{l_2\mathbf{k}_2} - s_{l_2l}^e \bar{C}_{l_1\mathbf{k}_1}^{lk})] \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}; \quad (\text{C8e})
\end{aligned}$$

(vi) equations for correlations \bar{D} involving hole operators and Mn position phase factors:

$$\begin{aligned}
\left(-i\hbar \frac{\partial}{\partial t} + E_{v_2\mathbf{k}_2} - E_{v_1\mathbf{k}_1} \right) \bar{D}_{v_1\mathbf{k}_1}^{v_2\mathbf{k}_2} &= \sum_l \mathbf{E} \cdot (\mathbf{M}_{v_2l-\mathbf{k}_2}^* (\bar{Y}_{v_1\mathbf{k}_1}^{l-\mathbf{k}_2})^* - \mathbf{M}_{v_1l-\mathbf{k}_1} \bar{Y}_{v_2\mathbf{k}_2}^{l-\mathbf{k}_1}) + \sum_v \mathbf{E} \cdot (\mathbf{M}_{v_2v\mathbf{k}_2}^h \bar{D}_{v_1\mathbf{k}_1}^{v_2\mathbf{k}_2} - \mathbf{M}_{v_1v\mathbf{k}_1}^h \bar{D}_{v_1\mathbf{k}_1}^{v_2\mathbf{k}_2}) \\
&+ J_{pd} \sum_{nn'v} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} [s_{v_1v}^h (n_{\text{Mn}} \bar{D}_{v\mathbf{k}_1}^{v_2\mathbf{k}_2} + D_{v_2\mathbf{k}_2}^{v_2}) - s_{v_2v}^h (n_{\text{Mn}} \bar{D}_{v_1\mathbf{k}_1}^{v_2\mathbf{k}_2} + D_{v_1\mathbf{k}_1}^v)] \right. \\
&\left. + \int_{\text{BZ}} [(s_{v_1v}^h \bar{K}_{D_{v\mathbf{k}_1}^{v_2n'\mathbf{k}_2}} - s_{v_2v}^h \bar{K}_{D_{v_1n\mathbf{k}_1}}^{vn'\mathbf{k}_1}) + M_n^{n'} (s_{v_1v}^h \bar{D}_{v\mathbf{k}_1}^{v_2\mathbf{k}_2} - s_{v_2v}^h \bar{D}_{v_1\mathbf{k}_1}^{v\mathbf{k}_1})] \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}; \quad (\text{C8f})
\end{aligned}$$

(vii) equations for correlations \bar{Y} involving electron-hole operators and Mn position phase factors:

$$\begin{aligned}
\left(-i\hbar \frac{\partial}{\partial t} + E_{v_1\mathbf{k}_1} + E_{l_2\mathbf{k}_2} \right) \bar{Y}_{v_1\mathbf{k}_1}^{l_2\mathbf{k}_2} &= \sum_l \mathbf{E} \cdot (\mathbf{M}_{l_2l\mathbf{k}_2}^e \bar{Y}_{v_1\mathbf{k}_1}^{l_2\mathbf{k}_2} - \mathbf{M}_{l_1v_1-\mathbf{k}_1} \bar{C}_{l_1-\mathbf{k}_1}^{l_2\mathbf{k}_2}) + \sum_v \mathbf{E} \cdot (\mathbf{M}_{v_1v\mathbf{k}_1}^h \bar{Y}_{v\mathbf{k}_1}^{l_2\mathbf{k}_2} - \mathbf{M}_{l_2v\mathbf{k}_2} \bar{D}_{v-\mathbf{k}_2}^{v_1\mathbf{k}_1}) \\
&- \sum_{nn'} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} \left[J_{pd} \sum_v s_{v\mathbf{k}_1}^h (n_{\text{Mn}} \bar{Y}_{v\mathbf{k}_1}^{l_2\mathbf{k}_2} + Y_{v-\mathbf{k}_2}^{l_2}) + J_{sd} \sum_l s_{l_2l}^e (n_{\text{Mn}} \bar{Y}_{v_1\mathbf{k}_1}^{l_2\mathbf{k}_2} + Y_{v_1\mathbf{k}_1}^l) \right] \right. \\
&\left. + \int_{\text{BZ}} \left[J_{pd} \sum_v s_{v\mathbf{k}_1}^h (\bar{K}_{Y_{v\mathbf{k}_1}^{l_2n'\mathbf{k}_2}} + M_n^{n'} \bar{Y}_{v\mathbf{k}_1}^{l_2\mathbf{k}_2}) + J_{sd} \sum_l s_{l_2l}^e (\bar{K}_{Y_{v_1n\mathbf{k}_1}^{ln'\mathbf{k}_1}} + M_n^{n'} \bar{Y}_{v_1\mathbf{k}_1}^{lk}) \right] \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}; \quad (\text{C8g})
\end{aligned}$$

(viii) equations for correlations \bar{K}_C involving electron and Mn spin operators as well as Mn position phase factors:

$$\left(-i\hbar \frac{\partial}{\partial t} + E_{l_2\mathbf{k}_2} - E_{l_1\mathbf{k}_1} \right) \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} = Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{E}} + Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{III}} + Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{IV}} + Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{V}}, \quad (\text{C8h})$$

where

$$\begin{aligned}
Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{E}} &:= \sum_v \mathbf{E} \cdot (\mathbf{M}_{v l_2\mathbf{k}_2}^* (\bar{K}_{Y_{v n_2-\mathbf{k}_2}}^{l_1 n_1 \mathbf{k}_1})^* - \mathbf{M}_{v l_1\mathbf{k}_1} \bar{K}_{Y_{v n_1-\mathbf{k}_1}}^{l_2 n_2 \mathbf{k}_2}) + \sum_l \mathbf{E} \cdot (\mathbf{M}_{l_2l\mathbf{k}_2}^e \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - \mathbf{M}_{l_1\mathbf{k}_1}^e \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}), \\
Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{III}} &:= J_{sd} \sum_{nl} \left\{ (\mathbf{S}_{n n_1} \cdot s_{l_1l}^e C_{l_2\mathbf{k}_2}^{l_2} M_n^{n_2} - \mathbf{S}_{n n_2} \cdot s_{l_2l}^e C_{l_1\mathbf{k}_1}^l M_n^{n_1}) \right. \\
&\left. + \int_{\text{BZ}} [\mathbf{S}_{n n_1} \cdot s_{l_1l}^e (\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} + \bar{C}_{l_2\mathbf{k}_2}^{l_2} M_n^{n_2}) - \mathbf{S}_{n n_2} \cdot s_{l_2l}^e (\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{ln\mathbf{k}_1} + \bar{C}_{l_1\mathbf{k}_1}^{lk} M_n^{n_1})] \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}, \\
Q_{\bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}}^{\text{IV}} &:= J_{sd} \sum_{nn'l} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} [n_{\text{Mn}} (s_{l_1l}^e \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - s_{l_2l}^e \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{ln_2\mathbf{k}_2}) - M_{n_1}^{n_2} (s_{l_1l}^e C_{l_2\mathbf{k}_2}^{l_2} - s_{l_2l}^e C_{l_1\mathbf{k}_1}^l)] \right. \\
&\left. - \frac{M_{n_1}^{n_2}}{(2\pi)^3} \int_{\text{BZ}} [(s_{l_1l}^e \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - s_{l_2l}^e \bar{K}_{C_{l_1n_1\mathbf{k}_1}}^{ln_2\mathbf{k}_2}) + M_n^{n'} (s_{l_1l}^e \bar{C}_{l_2\mathbf{k}_2}^{l_2} - s_{l_2l}^e \bar{C}_{l_1\mathbf{k}_1}^{lk})] d^3\mathbf{k} \right\},
\end{aligned}$$

$$\begin{aligned}
 Q_{\bar{K}C_{l_1n_1k_1}}^V &:= J_{pd} \sum_{nvv'} s_{vv'}^h \cdot \left\{ \frac{(Y_{v-k_1}^{l_1})^*}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_Y^{l_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_Y^{l_2n_2k_2}) d^3\mathbf{k} + Y_{v-k_1}^{l_2} (Y_{v-k_1}^{l_1})^* (\mathbf{S}_{nn_1} (M_n^{n_2})^* - \mathbf{S}_{n_2n} (M_n^{n_1})^*) \right. \\
 &+ \int_{\text{BZ}} [Y_{v-k_2}^{l_2} (\mathbf{S}_{nn_1} (\bar{K}_Y^{l_1n_2k_1})^* - \mathbf{S}_{n_2n} (\bar{K}_Y^{l_1n_2k_1})^*) + D_{v\mathbf{k}}^{v'} (\mathbf{S}_{nn_1} \bar{K}_{C_{l_1n_1k_1}}^{l_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_{C_{l_1n_1k_1}}^{l_2n_2k_2})] \frac{d^3\mathbf{k}}{(2\pi)^3} \\
 &+ \int_{\text{BZ}} ((Y_{v-k_1}^{l_1})^* \bar{Y}_{v\mathbf{k}}^{l_2k_2} + Y_{v-k_2}^{l_2} (\bar{Y}_{v\mathbf{k}}^{l_1k_1})^*) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1}) \frac{d^3\mathbf{k}}{(2\pi)^3} \\
 &- \frac{\bar{C}_{l_1k_1}^{l_2k_2}}{(2\pi)^6} \int \int_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{D_{v\mathbf{k}}}^{v'n_2k'} - \mathbf{S}_{n_2n} \bar{K}_{D_{v\mathbf{k}}}^{v'n_2k'}) + \bar{D}_{v\mathbf{k}}^{v'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1})] d^3\mathbf{k}' d^3\mathbf{k} \Big\} \\
 &- J_{sd} \sum_{nll'} s_{ll'}^e \cdot \left\{ \frac{C_{l_1k_1}^{l'}}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{C_{ln\mathbf{k}}}^{l_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_{C_{ln\mathbf{k}}}^{l_2n_2k_2}) d^3\mathbf{k} + C_{l_2k_2}^{l_2} C_{l_1k_1}^{l'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1}) \right\} \\
 &+ \int_{\text{BZ}} [C_{l_2k_2}^{l_2} (\mathbf{S}_{nn_1} \bar{K}_{C_{l_1n_1k_1}}^{l'n_2k} - \mathbf{S}_{n_2n} \bar{K}_{C_{l_1n_1k_1}}^{l'n_2k}) - C_{l_2k_2}^{l_2} (\mathbf{S}_{nn_1} \bar{K}_{C_{l_1n_1k_1}}^{l_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_{C_{l_1n_1k_1}}^{l_2n_2k_2})] \frac{d^3\mathbf{k}}{(2\pi)^3} \\
 &+ \int_{\text{BZ}} (C_{l_1k_1}^{l'} \bar{C}_{l_2k_2}^{l_2k_2} + C_{l_2k_2}^{l_2} \bar{C}_{l_1k_1}^{l_1k_1}) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1}) \frac{d^3\mathbf{k}}{(2\pi)^3} \\
 &+ \frac{\bar{C}_{l_1k_1}^{l_2k_2}}{(2\pi)^6} \int \int_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{C_{ln\mathbf{k}}}^{l'n_2k'} - \mathbf{S}_{n_2n} \bar{K}_{C_{ln\mathbf{k}}}^{l'n_2k'}) + \bar{C}_{l_2k_2}^{l_2k_2} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1})] d^3\mathbf{k}' d^3\mathbf{k} \Big\};
 \end{aligned}$$

(ix) equations for correlations \bar{K}_D involving hole and Mn spin operators as well as Mn position phase factors:

$$\left(-i\hbar \frac{\partial}{\partial t} + E_{v_2k_2} - E_{v_1k_1} \right) \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2} = Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^E + Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^{\text{III}} + Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^{\text{IV}} + Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^V, \quad (\text{C8i})$$

where

$$\begin{aligned}
 Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^E &:= \sum_l \mathbf{E} \cdot (\mathbf{M}_{v_2l-k_2}^* (\bar{K}_{Y_{v_1n_2k_1}}^{l_1n_1-k_2})^* - \mathbf{M}_{v_1l-k_1} \bar{K}_Y^{l_2n_2-k_1}) + \sum_v \mathbf{E} \cdot (\mathbf{M}_{v_2v_2k_2}^h \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2} - \mathbf{M}_{v_1v_1k_1}^h \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}), \\
 Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^{\text{III}} &:= J_{pd} \sum_{nv} \left\{ (\mathbf{S}_{nn_1} \cdot s_{vv'}^h D_{v_2k_2}^{v_2} M_n^{n_2} - \mathbf{S}_{n_2n} \cdot s_{v_2v}^h D_{v_1k_1}^v M_n^{n_1}) \right. \\
 &+ \int_{\text{BZ}} \left[\mathbf{S}_{nn_1} \cdot s_{vv_1}^h (\bar{K}_{D_{v\mathbf{k}}}^{v_2n_2k_2} + \bar{D}_{v\mathbf{k}}^{v_2k_2} M_n^{n_2}) - \mathbf{S}_{n_2n} \cdot s_{v_2v}^h (\bar{K}_{D_{v_1n_1k_1}}^{v\mathbf{k}} + \bar{D}_{v_1k_1}^v M_n^{n_1}) \right] \frac{d^3\mathbf{k}}{(2\pi)^3} \Big\}, \\
 Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^{\text{IV}} &:= J_{pd} \sum_{nn'v} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} [n_{\text{Mn}} (s_{vv_1}^h \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2} - s_{v_2v}^h \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}) - M_n^{n_2} (s_{vv_1}^h D_{v_2k_2}^{v_2} - s_{v_2v}^h D_{v_1k_1}^v)] \right. \\
 &- \frac{M_n^{n_2}}{(2\pi)^3} \int_{\text{BZ}} [(s_{vv_1}^h \bar{K}_{D_{v\mathbf{k}}}^{v_2n_2k_2} - s_{v_2v}^h \bar{K}_{D_{v_1n_1k_1}}^{v\mathbf{k}}) + M_n^{n'} (s_{vv_1}^h \bar{D}_{v\mathbf{k}}^{v_2k_2} - s_{v_2v}^h \bar{D}_{v_1k_1}^v)] d^3\mathbf{k} \Big\}, \\
 Q_{\bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2}}^V &:= J_{sd} \sum_{nll'} s_{ll'}^e \cdot \left\{ \frac{(Y_{v_1k_1}^{l_1})^*}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_Y^{l'n_2k} - \mathbf{S}_{n_2n} \bar{K}_Y^{l'n_2k}) d^3\mathbf{k} + Y_{v_2k_2}^{l_2} (Y_{v_1k_1}^{l_1})^* (\mathbf{S}_{nn_1} (M_n^{n_2})^* - \mathbf{S}_{n_2n} (M_n^{n_1})^*) \right. \\
 &+ \int_{\text{BZ}} [Y_{v_2k_2}^{l_2} (\mathbf{S}_{nn_1} (\bar{K}_Y^{l'n_2k})^* - \mathbf{S}_{n_2n} (\bar{K}_Y^{l'n_2k})^*) + C_{l_2k_2}^{l_2} (\mathbf{S}_{nn_1} \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2})] \frac{d^3\mathbf{k}}{(2\pi)^3} \\
 &+ \int_{\text{BZ}} ((Y_{v_1k_1}^{l_1})^* \bar{Y}_{v_2k_2}^{l_2k_2} + Y_{v_2k_2}^{l_2} (\bar{Y}_{v_1k_1}^{l_1k_1})^*) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1}) \frac{d^3\mathbf{k}}{(2\pi)^3} \\
 &- \frac{\bar{D}_{v_1k_1}^{v_2k_2}}{(2\pi)^6} \int \int_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{C_{ln\mathbf{k}}}^{l'n_2k'} - \mathbf{S}_{n_2n} \bar{K}_{C_{ln\mathbf{k}}}^{l'n_2k'}) + \bar{C}_{l_2k_2}^{l_2k_2} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1})] d^3\mathbf{k}' d^3\mathbf{k} \Big\} \\
 &- J_{pd} \sum_{nvv'} s_{vv'}^h \cdot \left\{ \frac{D_{v_1k_1}^{v'}}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{D_{v\mathbf{k}}}^{v_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_{D_{v\mathbf{k}}}^{v_2n_2k_2}) d^3\mathbf{k} + D_{v_2k_2}^{v_2} D_{v_1k_1}^{v'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_n^{n_1}) \right. \\
 &+ \int_{\text{BZ}} [D_{v_2k_2}^{v_2} (\mathbf{S}_{nn_1} \bar{K}_{D_{v_1n_1k_1}}^{v'n_2k} - \mathbf{S}_{n_2n} \bar{K}_{D_{v_1n_1k_1}}^{v'n_2k}) - D_{v\mathbf{k}}^{v'} (\mathbf{S}_{nn_1} \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2} - \mathbf{S}_{n_2n} \bar{K}_{D_{v_1n_1k_1}}^{v_2n_2k_2})] \frac{d^3\mathbf{k}}{(2\pi)^3}
 \end{aligned}$$

$$\begin{aligned}
& + \int_{\text{BZ}} (D_{v_1\mathbf{k}_1}^{v'} \bar{D}_{v\mathbf{k}}^{v_2\mathbf{k}_2} + D_{v\mathbf{k}_2}^{v_2} \bar{D}_{v_1\mathbf{k}_1}^{v'\mathbf{k}'}) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) \frac{d^3\mathbf{k}}{(2\pi)^3} \\
& + \frac{\bar{D}_{v_1\mathbf{k}_1}^{v_2\mathbf{k}_2}}{(2\pi)^6} \int \int_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{D_{v\mathbf{k}}}^{v'n_2\mathbf{k}'} - \mathbf{S}_{n_2n} \bar{K}_{D_{v_1\mathbf{k}_1}}^{v'n\mathbf{k}'}) + \bar{D}_{v\mathbf{k}}^{v'\mathbf{k}'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n)] d^3\mathbf{k}' d^3\mathbf{k} \Big\};
\end{aligned}$$

(x) equations for correlations \bar{K}_Y involving electron-hole and Mn spin operators as well as Mn position phase factors:

$$\left(-i\hbar \frac{\partial}{\partial t} + E_{v_1\mathbf{k}_1} + E_{l_2\mathbf{k}_2}\right) \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} = Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{E}} + Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{III}} + Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{IV}} + Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{V}}, \quad (\text{C8j})$$

where

$$\begin{aligned}
Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{E}} & := \sum_l \mathbf{E} \cdot (\mathbf{M}_{l_2/l\mathbf{k}_2}^e \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - \mathbf{M}_{l_{v_1}-\mathbf{k}_1} \bar{K}_{C_{l_{n_1}-\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}) + \sum_v \mathbf{E} \cdot (\mathbf{M}_{v_1v\mathbf{k}_1}^h \bar{K}_{Y_{vn_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - \mathbf{M}_{l_2v\mathbf{k}_2} \bar{K}_{D_{vn_1-\mathbf{k}_2}}^{v_1n_2\mathbf{k}_1}), \\
Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{III}} & := - \sum_n \mathbf{S}_{n_2n} \cdot \left\{ \left(J_{sd} \sum_l Y_{v_1\mathbf{k}_1}^l M_{n_1}^n s_{l_2l}^e + J_{pd} \sum_v Y_{v-\mathbf{k}_2}^{l_2} M_{n_1}^n s_{v_1v}^h \right) \right. \\
& \quad \left. + \int_{\text{BZ}} \left[J_{sd} \sum_l (\bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{ln\mathbf{k}} + \bar{Y}_{v_1\mathbf{k}_1}^{l\mathbf{k}} M_{n_1}^n) s_{l_2l}^e + J_{pd} \sum_v (\bar{K}_{Y_{vn_1\mathbf{k}_1}}^{l_2n\mathbf{k}_2} + \bar{Y}_{v\mathbf{k}}^{l_2\mathbf{k}_2} M_{n_1}^n) s_{v_1v}^h \right] \frac{d^3\mathbf{k}}{(2\pi)^3} \right\}, \\
Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{IV}} & := - \sum_{nn'} \mathbf{S}_{nn'} \cdot \left\{ M_n^{n'} \left[n_{\text{Mn}} \left(J_{pd} \sum_v s_{v_1v}^h \bar{K}_{Y_{vn_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} + J_{sd} \sum_l s_{l_2l}^e \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} \right) \right. \right. \\
& \quad \left. \left. - M_{n_1}^{n_2} \left(J_{pd} \sum_v s_{v_1v}^h Y_{v-\mathbf{k}_2}^{l_2} + J_{sd} \sum_l s_{l_2l}^e Y_{v_1\mathbf{k}_1}^l \right) \right] \right. \\
& \quad \left. - \frac{M_{n_1}^{n_2}}{(2\pi)^3} \int_{\text{BZ}} \left[\left(J_{pd} \sum_v s_{v_1v}^h \bar{K}_{Y_{v\mathbf{k}}}^{l_2n'\mathbf{k}_2} + J_{sd} \sum_l s_{l_2l}^e \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n'\mathbf{k}_2} \right) + M_n^{n'} \left(J_{pd} \sum_v s_{v_1v}^h \bar{Y}_{v\mathbf{k}}^{l_2\mathbf{k}_2} + J_{sd} \sum_l s_{l_2l}^e \bar{Y}_{v_1\mathbf{k}_1}^{l_2\mathbf{k}_2} \right) \right] d^3\mathbf{k} \right\}, \\
Q_{\bar{K}_Y^{l_2n_2\mathbf{k}_2}}^{\text{V}} & := - J_{sd} \sum_{nll'} s_{ll'}^e \cdot \left\{ \frac{C_{lk_2}^{l_2}}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l'n_2\mathbf{k}} - \mathbf{S}_{n_2n} \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l'n\mathbf{k}}) d^3\mathbf{k} + Y_{v_1\mathbf{k}_1}^{l'} C_{lk_2}^{l_2} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) \right. \\
& \quad \left. + \frac{1}{(2\pi)^3} \int_{\text{BZ}} \left[Y_{v_1\mathbf{k}_1}^{l'} (\mathbf{S}_{nn_1} \bar{K}_{C_{l\mathbf{k}}}^{l_2n_2\mathbf{k}_2} - \mathbf{S}_{n_2n} \bar{K}_{C_{l\mathbf{k}}}^{l_2n_2\mathbf{k}_2}) - C_{l\mathbf{k}}^{l'} (\mathbf{S}_{nn_1} \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - \mathbf{S}_{n_2n} \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2}) \right] d^3\mathbf{k} \right. \\
& \quad \left. + \int_{\text{BZ}} (C_{lk_2}^{l_2} \bar{Y}_{v_1\mathbf{k}_1}^{l'\mathbf{k}} + Y_{v_1\mathbf{k}_1}^{l'} \bar{C}_{l\mathbf{k}}^{l_2\mathbf{k}_2}) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) \frac{d^3\mathbf{k}}{(2\pi)^3} \right. \\
& \quad \left. + \frac{\bar{Y}_{v_1\mathbf{k}_1}^{l_2\mathbf{k}_2}}{(2\pi)^6} \int \int_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{C_{l\mathbf{k}}}^{l'n_2\mathbf{k}'} - \mathbf{S}_{n_2n} \bar{K}_{C_{l_{n_1}\mathbf{k}}}^{l'n\mathbf{k}'}) + \bar{C}_{l\mathbf{k}}^{l'\mathbf{k}'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n)] d^3\mathbf{k}' d^3\mathbf{k} \right\} \\
& \quad - J_{pd} \sum_{nvv'} s_{vv'}^h \cdot \left\{ \frac{D_{v\mathbf{k}_1}^{v_1}}{(2\pi)^3} \int_{\text{BZ}} (\mathbf{S}_{nn_1} \bar{K}_{Y_{v'\mathbf{k}}}^{l_2n_2\mathbf{k}_2} - \mathbf{S}_{n_2n} \bar{K}_{Y_{v'\mathbf{k}}}^{l_2n_2\mathbf{k}_2}) d^3\mathbf{k} + Y_{v'-\mathbf{k}_2}^{l_2} D_{v\mathbf{k}_1}^{v_1} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) \right. \\
& \quad \left. + \frac{1}{(2\pi)^3} \int_{\text{BZ}} [Y_{v'-\mathbf{k}_2}^{l_2} (\mathbf{S}_{nn_1} \bar{K}_{D_{v\mathbf{k}}}^{v_1n_2\mathbf{k}_1} - \mathbf{S}_{n_2n} \bar{K}_{D_{v_1\mathbf{k}_1}}^{v_1n\mathbf{k}_1}) - D_{v\mathbf{k}}^{v'} (\mathbf{S}_{nn_1} \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2} - \mathbf{S}_{n_2n} \bar{K}_{Y_{v_1n_1\mathbf{k}_1}}^{l_2n_2\mathbf{k}_2})] d^3\mathbf{k} \right. \\
& \quad \left. + \int_{\text{BZ}} (D_{v\mathbf{k}_1}^{v_1} \bar{Y}_{v'\mathbf{k}}^{l_2\mathbf{k}_2} + Y_{v'-\mathbf{k}_2}^{l_2} \bar{D}_{v\mathbf{k}}^{v_1\mathbf{k}_1}) (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n) \frac{d^3\mathbf{k}}{(2\pi)^3} \right. \\
& \quad \left. + \frac{\bar{Y}_{v_1\mathbf{k}_1}^{l_2\mathbf{k}_2}}{(2\pi)^6} \int \int_{\text{BZ}} [(\mathbf{S}_{nn_1} \bar{K}_{D_{v\mathbf{k}}}^{v'n_2\mathbf{k}'} - \mathbf{S}_{n_2n} \bar{K}_{D_{v_1\mathbf{k}_1}}^{v'n\mathbf{k}'}) + \bar{D}_{v\mathbf{k}}^{v'\mathbf{k}'} (\mathbf{S}_{nn_1} M_n^{n_2} - \mathbf{S}_{n_2n} M_{n_1}^n)] d^3\mathbf{k}' d^3\mathbf{k} \right\}.
\end{aligned}$$

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