Magnetoresistance in organic semiconductors: Including pair correlations in the kinetic equations for hopping transport

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We derive kinetic equations for polaron hopping in organic materials that explicitly take into account the double occupation possibility and pair intersite correlations. The equations include simplified phenomenological spin dynamics and provide a self-consistent framework for the description of the bipolaron mechanism of the organic magnetoresistance. At low applied voltages, the equations can be reduced to those for an effective resistor network that generalizes the Miller-Abrahams network and includes the effect of spin relaxation on the system resistivity. Our theory discloses the close relationship between the organic magnetoresistance and the intersite correlations. Moreover, in the absence of correlations, as in an ordered system with zero Hubbard energy, the magnetoresistance vanishes.

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I. INTRODUCTION

The transport properties of organic materials represent an interesting and fast-developing research field. While a number of basic properties, especially those related to light emitting devices [1] and field effect transistors [2], have been well understood, many open issues are still under debate. Among these, deep and complex issues on the transport of spin-polarized carriers or magnetoresistance effects in organic materials have been risen recently by the advent of molecular spintronics [3].

Transport in organic materials is usually described as a hopping conductivity promoted by polaron hops, and its theoretical description is to some extent similar to the conventional theory of hopping conduction [4]. However, this transport is also characterized by a number of features not acknowledged in classical systems with hopping. One of this features is the so-called organic magnetoresistance (OMAR), a strong magnetoresistance easily detectable in relatively weak magnetic fields of ∼10–100 Gs. The qualitative understanding of this effect [5–7] is based on the simple observation that the spin relaxation influences the hopping transport, as for example demonstrated by Monte-Carlo simulations [7]. Nevertheless, important basic issues, and especially the role of correlations, remain so far unclear.

The conventional description of the hopping conduction is based on the Miller-Abrahams network. This network and the underlying kinetic equations can be derived from quantum mechanics, while its applicability is limited by several constrains. One of the most important constraint is the so-called Hartree decoupling that requires the exclusion of intersite correlations. The average of the product of two filling numbers $\overline{n_i n_j}$ is considered to be equal to the product of averaged filling numbers $\overline{n}_i \overline{n}_j$ [8]. This condition is satisfied in the equilibrium when the Coulomb interaction between charge carriers is not taken into account.

The Coulomb interaction induces, nevertheless, strong intersite correlations, leading, for example, to the phenomenon of the Coulomb glass [9–11]. Moreover, the intersite correlations can appear even without the Coulomb interaction when the system is out of the equilibrium. Actually, the applied voltage that induces electric current drives the system out of equilibrium and can lead to these correlations. Recently, it was shown [12,13] that the correlations can affect the transport properties of the system even in the linear-response regime.

Here, we discuss the correlations in the context of organic magnetoresistance. Specifically, we discuss the theory of the bipolaron mechanism of OMAR and show how spin relaxation renormalizes the hopping transport via correlations. The bipolaron mechanism was first proposed in Ref. [7] in terms of a smart theoretical model based on the parallel and antiparallel configurations of spins on two sites. The effect was demonstrated with the Monte Carlo simulations. This study was followed by several attempts to include these parallel/antiparallel configurations into the conventional theory of hopping conduction [14–20]. The number of these attempts itself indicate the interest in generalizing the conventional theory of hopping transport to include effects similar to OMAR. However, all these attempts faced one problem. The conventional approach [4] to the percolation theory follows the scheme: the rate equations are derived first from the quantum mechanics with the Hartree decoupling, then the resistor network is obtained as a linearization of these equations. Only after that the percolation theory is applied to describe the resistivity of this network. However, we showed recently [21] that kinetic equations with Hartree decoupling cannot describe the organic magnetoresistance. The mentioned studies [14–17,19,20] did not provide a rederivation of the theory. Rather the bipolaron qualitative mechanism of OMAR was artificially included in the conventional percolation picture excluding the intersite correlations. In the present study we show that these correlations naturally include the spin relaxation and are fundamental for the description of the OMAR. Note that the Monte-Carlo simulations performed in Ref. [7] automatically include all intersite correlations that are neglected in the Hartree decoupling.

Although we recognize the importance of previous models that fused OMAR with percolation theory we expand the conventional approach by explicitly including the intercite correlations. Namely, we derive the kinetic equations that take into account pair intersite correlations and the possibility of the double occupation. The equations also include spin dynamics in the simplified phenomenological model where it is described by a single spin relaxation time dependent on the magnetic field. We demonstrate that at low applied voltage these equations can be reduced to an effective resistor network. The expression for the effective resistance between sites i and j is more complex than in Miller-Abrahams theory and is dependent on the other sites surrounding the pair ij. However, this expression depends explicitly on the spin degrees of freedom and can describe the organic magnetoresistance.

The paper is organized as follows. In Sec. II, we give a short qualitative review of the bipolaron mechanism of OMAR. In Sec. III, we derive the kinetic equations that include pair intersite correlations in the simple case of large Hubbard energy. We also provide the equations in the general case of arbitrary Hubbard energy. The derivation for the general case is presented in Supplementary Material [22]. In Sec. IV, we linearize the kinetic equations to obtain the effective resistor network. The obtained network generalizes the Miller-Abrahams resistor network by including pair intersite correlations. It can be used to construct the rigorous percolation theory that describes OMAR. In Sec. V, we study the kinetic equations numerically and discuss the main features of OMAR that follow from these equations. In Sec. VI, we provide the general discussion of the obtained results.

II. BIPOLARON MECHANISM OF OMAR

It was proposed [7] that the organic magnetoresistance can be described with the bipolaron mechanism. The main idea of the mechanism is the so-called "spin blocking." It requires the possibility of double occupation of the hopping site. It means that an electron can hop to a site already occupied with one electron and form a bipolaron provided that the spins of both electrons form a singlet. However, the two electrons with the same spin projections along the common quantization axis have zero singlet probability and cannot form a bipolaron. This leads to effective reduction of the number of sites available for hopping. The spin relaxation can rotate the electron spins, change the parallel configuration to antiparallel, and therefore restore the possibility of the hop. Note that the magnetic field corresponding to organic magnetoresistance is weak and Zeeman energy is much smaller than temperature. Therefore spin polarization is absent and the number of polarons with up and down spin projections is always the same. However, the spin dynamics still influence the current, because the magnetic field alters the spin relaxation process.

One of the dominant mechanisms of the spin relaxation in organic materials is the hyperfine interaction with nuclear magnetic moments. The applied magnetic field ~10–100 Gs influences significantly this mechanism. The nature of the effect of the applied magnetic field on the spin relaxation rate is shown on Fig. 1. The mutual orientation of the spins of electrons on sites i and j relaxes because the spins rotate around the different local hyperfine fields $\mathbf{H}_{\rm HF}(i)$ and $\mathbf{H}_{\rm HF}(j)$ [Fig. 1(a)]. When the applied magnetic field is larger than the hyperfine field [Fig. 1(b)] the spins rotate around the total field $\mathbf{H}+\mathbf{H}_{\rm HF}$. This total field differs only slightly on sites i and j. It significantly suppress the spin relaxation.

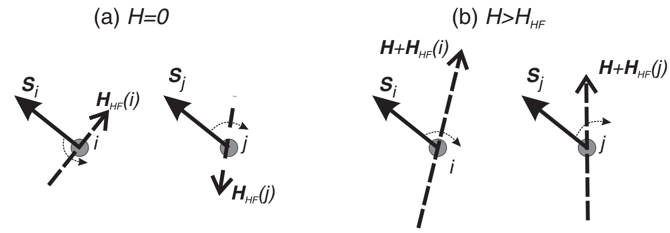

FIG. 1. The effect of the applied magnetic field on the hyperfine mechanism of spin relaxation.

The rigorous theoretical description of the mechanism of spin relaxation is quite complex and includes interplay between the polaron hopping and the on-site spin rotation [23–26]. It leads to many sophisticated phenomena such as the slow nonexponential tails in the spin relaxation [21,26]. In the present study, we are not specifically interested in the details of the spin relaxation. Rather we focus on the question why the spin relaxation affects the charge transport in the situation when the spin polarization is absent. Therefore we adopt an oversimplified model that reduces the complex physics of the hyperfine mechanism of spin relaxation to the single spin-flip rate τ_s^{-1} that is dependent on the applied magnetic field.

For this rate, we use the expression

$$\tau_s^{-1} = \omega_s^{(0)} \frac{H_{\rm HF}^2}{H_{\rm HF}^2 + H^2}. \tag{1}$$

Here, $\omega_s^{(0)}$ is the zero magnetic field spin-flip rate. $H_{\rm HF}$ is the typical value of the hyperfine field and H is the applied magnetic field. The term $H_{\rm HF}^2/(H^2+H_{\rm HF}^2)$ reflects the suppression of the spin relaxation in the applied magnetic field. Note that the reduction of the hyperfine spin relaxation to the magnetic field dependent spin-flip rate was used in several studies describing the organic magnetoresistance. Equation (1) agrees with the approach used in Refs. [14,17]

III. KINETIC EQUATIONS WITH INTERSITE PAIR CORRELATIONS

Conventionally, the derivation of the theory of hopping conduction starts from kinetic equations

$$\frac{d\overline{n}_{i,\uparrow}}{dt} = \sum_j W_{ji}\overline{n_{j,\uparrow}p_{i,\uparrow}} - W_{ij}\overline{n_{i,\uparrow}p_{j,\uparrow}}. \tag{2}$$

Here, $\overline{n}_{i,\uparrow}$ is the average filling number of spin-up electron on site i. $p_{j,\uparrow}$ stands for the possibility to find an empty place for spin-up electron on site j. The mutual line over $n_{i,\uparrow}$ and $p_{j,\uparrow}$ corresponds to the joint averaging of the product $n_{i,\uparrow}p_{j,\uparrow}$.

The next conventional step that impedes the description of the organic magnetoresistance is the Hartree decoupling.

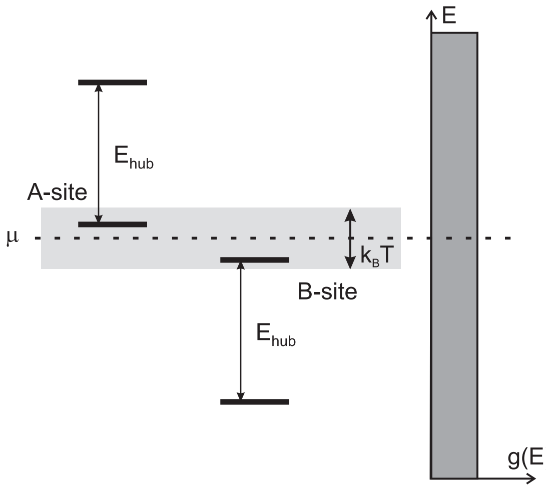

FIG. 2. The cartoon for A-type and B-type sites. A-type sites have the single occupation states in the energy band of width $k_B T$ around the Fermi level (shown with light grey color). B-type sites have the double-occupied states in this band. To provide both types of sites, the density of states $g(E)$ should be wider than E_{hub}.

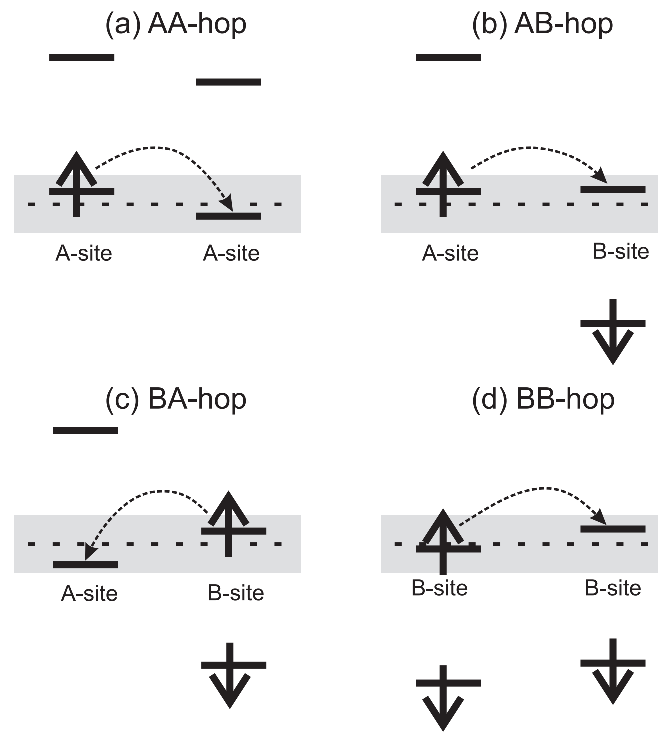

FIG. 3. Hopping between different kinds of sites. The AB hop (b) is allowed only for opposite spin directions of the hopping electron and of the electron on "free" B-type site. Other hops are always possible. The BA hop (c) always results in an antiparallel spin configuration on the sites.

This approximation corresponds to the substitution $\overline{n_{i,\uparrow} p_{j,\uparrow}} \rightarrow \overline{n}_{i,\uparrow} \overline{p}_{j,\uparrow}$. It allows us to obtain many important physical results, for example, the temperature dependence of conductivity and orbital magnetoresistance [4] but is known to be insufficient for the description of OMAR [21]. Here, we go one step beyond this approximation. We include pair correlations of the filling numbers but exclude triple correlations. It means that when we consider a pair of sites i-j we make the decoupling $\overline{n_{i\uparrow} n_{j\uparrow} n_{k\uparrow}} \rightarrow \overline{n_{i\uparrow} n_{j\uparrow}} \cdot \overline{n}_{k\uparrow}$. Our assumptions are similar to the approach used recently in Refs. [12,13]. However, the theory [12,13] does not include the double occupation possibility that is essential for the bipolaron mechanism of OMAR.

We start our consideration from the simplest model that describes hopping with double occupation possibility. This model assumes that the Hubbard energy E_{hub} is much larger than temperature, but the localization sites have a broad energy distribution $g(E)$ with a width larger than E_{hub}. In order to effectively participate in the hopping, the localization site should have a state with the energy close to the chemical potential μ. The states far below μ are always filled and the states with too large energies are never occupied. Only the sites with an energy level in some energy band with width $\sim k_B T$ around the chemical potential can change their filling numbers and effectively participate in hopping. This band is shown by a light grey color in Fig. 2. More rigorously, in the situation with an exponentially broad distribution of the hopping rates, the width of the band is $\xi_c k_B T$, where ξ_c is the percolation exponent [4]. The condition of large Hubbard energy in this case reads $E_{\text{hub}} \gg \xi_c k_B T$ but we limit our analysis to the case $\xi_c \sim 1$. For these conditions there are two types of sites that are important for conductivity (see Fig. 2) [27]. The so-called A-type sites have their energy E_i near the Fermi energy $E_i \sim \mu$. These sites are never double occupied because $E_i + E_{\text{hub}} - \mu \gg k_B T$. The B-type sites are characterized by single-occupation energy well below the chemical potential and these sites always have at least one electron. The energy of doubly occupied B-site is near the chemical potential $E_i + E_{\text{hub}} \sim \mu$, therefore these sites can have one or two electrons. The exact form of the density of states $g(E)$ is not very important for our study. In Fig. 2, we consider $g(E) = \text{const}$ in some interval of energies. Nevertheless, our consideration is valid for other $g(E)$ that allows the existence of both A-type and B-type sites. For example, it is valid for the model of host and donor states in polymers considered in Ref. [28].

In our model we consider small magnetic fields that cannot lead to the spin polarization. Therefore the averaged filling numbers of a site for different spin projections are always the same $\overline{n}_{i\uparrow} = \overline{n}_{i\downarrow}$. Also, we assume the spin inversion symmetry that leads to the conservation of any averaged value after inversion of all the spin projections, for example, $\overline{n_{i\uparrow} n_{j\downarrow}} = \overline{n_{i\downarrow} n_{j\uparrow}}$. However, this symmetry still allows the existence of the spin correlations. For A-site i and B-site j, the spin correlation can be defined as $s_{ij} = \overline{n_{i\uparrow} p_{j\uparrow}} - \overline{n_{i\uparrow} p_{j\downarrow}}$. Beside the spin correlations s_{ij}, the model allows another kind of pair correlations that are less dependent on the spin degree of freedom. We call them charge correlations c_{ij}. For A-B pair of sites i-j, it can be defined as $c_{ij} = \overline{n_{i\uparrow} p_{j\uparrow}} + \overline{n_{i\uparrow} p_{j\downarrow}} - 2\overline{n}_{i\uparrow} \overline{p}_{j\uparrow}$.

To describe these correlations, we should write not only the rate equations for averaged filling numbers $\overline{n}_{i\uparrow}$ but also the equations for their products, for example, for $\overline{n_{i\uparrow} p_{j\uparrow}}$. Let us assume that the site i is of type A and site j is of type B. In this case, $\overline{n_{i\uparrow} p_{j\uparrow}}$ stands for the probability for both sites to have one electron [this situation is shown in Fig. 3(b)]. The electron on site i has spin projection $\uparrow$ and the electron on site j has spin projection $\downarrow$ (it allows the hop of $\uparrow$ electron to the

B-type site j). The configuration $n_{i\uparrow}p_{j\uparrow}$ can appear due to the electron hopping in several different ways. First, it can appear due to the hop of spin-up electron from the site j to site i [see Fig. 3(c)]. Second, it can appear due to the hop of spin-up electron from the third site k to site i while the site j already has only one spin-down electron. Third, it can appear due to the hop of spin-up electron from site j to the third site m, while the site i is occupied by another spin-up electron. There are also three ways to break the configuration $n_{i\uparrow}p_{j\uparrow}$: the hop $i \to j$ and the hops of spin-up electron $i \to k$ or $m \to j$. Finally, the finite spin relaxation time τ_s leads to another possibility of the appearance and the disappearance of the configuration $n_{i\uparrow}p_{j\uparrow}$ due to the spin flip. Taking into account all the possibilities, we write the rate equation for $\overline{n_{i\uparrow}p_{j\uparrow}}$:

$$\begin{aligned}\frac{d\overline{n_{i\uparrow}p_{j\uparrow}}}{dt} &= W_{ji}\overline{p_{i\uparrow}n_{j\uparrow}} + \sum_{k\neq i,j} W_{ki}\overline{n}_{k\uparrow}\overline{p_{i\uparrow}p_{j\uparrow}} \\ &\quad + \sum_{m\neq i,j} W_{jm}\overline{p}_{m\uparrow}\overline{n_{i\uparrow}n_{j\uparrow}} + \frac{1}{\tau_s}\overline{n_{i\uparrow}p_{j\downarrow}} \\ &\quad - \overline{n_{i\uparrow}p_{j\uparrow}}\left(\frac{1}{\tau_s} + W_{ij} + \sum_{k\neq i,j} W_{ik}\overline{p}_{k\uparrow} + \sum_{m\neq i,j} W_{mj}\overline{n}_{m\uparrow}\right).\end{aligned} \tag{3}$$

In this equation, we took into account that $\overline{n_{i\uparrow}p_{j\downarrow}} = \overline{n_{i\downarrow}p_{j\uparrow}}$ due to the spin inversion symmetry. The values $n_{i\uparrow}$ and $p_{i\downarrow}$ are, of course, not independent. However, their relation is different for A-type and B-type sites. For A-type site i, $p_{i\uparrow}$ corresponds to the situation when the site i has no electrons. Therefore we can substitute $p_{i\uparrow} = p_{i\downarrow} = 1 - n_{i\uparrow} - n_{i\downarrow}$. We can make this substitution under the joint averaging or when all the terms are averaged separately. On the other hand, the substitution $n_{i\uparrow} = n_{i\downarrow}$ is possible only for separate averaging but not under the joint averaging. It means that $\overline{n}_{i\uparrow} = \overline{n}_{i\downarrow}$ but $\overline{n_{i\uparrow}p_{j\uparrow}} \neq \overline{n_{i\downarrow}p_{j\uparrow}}$. For the B-type site j, the value $n_{j\uparrow}$ corresponds to the situation when the site j is double-occupied and the substitution $n_{j\uparrow} = n_{j\downarrow} = 1 - p_{j\uparrow} - p_{j\downarrow}$ can be made both under the joint or the separate averaging of the terms. The substitution $p_{j\uparrow} \to p_{j\downarrow}$ is not possible under the joint averaging.

In the expression (3), we used the decoupling $\overline{n_{i\uparrow}n_{j\uparrow}p_{m\uparrow}} \to \overline{n_{i\uparrow}n_{j\uparrow}}\,\overline{p}_{m\uparrow}$. Strictly speaking, the correct decoupling taking into account all the pair correlations and neglecting triple correlations is $\overline{n_{i\uparrow}n_{j\uparrow}p_{m\uparrow}} \to \overline{n_{i\uparrow}n_{j\uparrow}}\,\overline{p}_{m\uparrow} + \overline{n}_{i\uparrow}\overline{n_{j\uparrow}p_{m\uparrow}} + \overline{n}_{j\uparrow}\overline{n_{i\uparrow}p_{m\uparrow}} - 2\overline{n}_{j\uparrow}\overline{n}_{i\uparrow}\overline{p}_{m\uparrow}$. This substitution becomes more clear if we express the averaged product $\overline{n_{i\uparrow}n_{j\uparrow}p_{m\uparrow}}$ in terms of product of averaged filling numbers $\overline{n}_{j\uparrow}\overline{n}_{i\uparrow}\overline{p}_{m\uparrow}$, double and triple correlations, and neglect the triple correlation. Here, we neglect "long-range" correlations. If site m is connected by the hopping with site j, we consider the hopping i-m to be irrelevant for the dynamics of correlations i-j. In this case, we neglect the correlations i-m. This assumption is similar to the one made in Refs. [12,13]. With this assumption, when we go from the averaged joint products to the correlations s_{ij} and c_{ij}, the additional terms corresponding to the pair correlations of sites i and j with other sites will be canceled. Therefore we do not write them from the beginning.

We believe that it is instructive to exclude p_i and n_j from the expression (3) to keep only the electron notations for the A-type site i and hole notations for B-type site j. However, we will keep both p_k and n_k for sites k and m because these sites play only auxiliary role in the determination of the correlations s_{ij} and c_{ij} and we are not particularly interested in their type,

$$\begin{aligned}\frac{d\overline{n_{i\uparrow}p_{j\uparrow}}}{dt} &= \frac{1}{\tau_s}\overline{n_{i\uparrow}p_{j\downarrow}} + W_{ji}(1 - 2\overline{n}_{i\uparrow} - 2\overline{p}_{j\uparrow} + 2\overline{n_{i\uparrow}p_{j\uparrow}} \\ &\quad + 2\overline{n_{i\uparrow}p_{j\downarrow}}) + \sum_{k\neq i,j} W_{ki}\overline{n}_{k\uparrow}(\overline{p}_{j\uparrow} - \overline{n_{i\uparrow}p_{j\uparrow}} - \overline{n_{i\uparrow}p_{j\downarrow}}) \\ &\quad + \sum_{m\neq i,j} W_{jm}\overline{p}_{m\uparrow}(\overline{n}_{i\uparrow} - \overline{n_{i\uparrow}p_{j\uparrow}} - \overline{n_{i\uparrow}p_{j\downarrow}}) \\ &\quad - \overline{n_{i\uparrow}p_{j\uparrow}}\left(\frac{1}{\tau_s} + W_{ij} + \sum_{k\neq i,j} W_{ik}\overline{p}_{k\uparrow} + \sum_{m\neq i,j} W_{mj}\overline{n}_{m\uparrow}\right).\end{aligned} \tag{4}$$

To get the equations for correlations s_{ij} and c_{ij}, it is necessary to write a similar equation for $\overline{n_{i\uparrow}p_{j\downarrow}}$ and combine it with Eq. (4) and with the rate equations for averaged filling numbers $\overline{n}_{i\uparrow}$ and $\overline{p}_{j\uparrow}$. The straightforward algebra yields

$$\frac{ds_{ij}}{dt} = J_{ji} - r_{s,ij}s_{ij}, \quad r_{s,ij} = \frac{1}{\tau_s} + r^{(0)}_{s,ij},$$

$$r^{(0)}_{s,ij} = \sum_{k\neq i,j} W_{ik}\overline{p}_{k\uparrow} + \sum_{m\neq i,j} W_{mj}\overline{n}_{m\uparrow}, \tag{5}$$

$$\frac{dc_{ij}}{dt} = J_{ji}(1 - 2\overline{n}_{i\uparrow} - 2\overline{p}_{j\uparrow}) - r_{c,ij}c_{ij}, \tag{6}$$

$$\begin{aligned}r_{c,ij} &= \sum_{k\neq i,j}(2W_{ki}\overline{n}_{k\uparrow} + W_{ik}\overline{p}_{k\uparrow}) \\ &\quad + \sum_{m\neq i,j}(2W_{jm}\overline{p}_{m\uparrow} + W_{mj}\overline{n}_{m\uparrow}).\end{aligned} \tag{7}$$

In this equations, $J_{ji} = W_{ji}(1 - 2\overline{n}_{i\uparrow} - 2\overline{p}_{j\uparrow} + 2\overline{n_{i\uparrow}p_{j\uparrow}} + 2\overline{n_{i\uparrow}p_{j\downarrow}}) - W_{ij}\overline{n_{i\uparrow}p_{j\uparrow}}$ is the current between sites j and i carried by spin-up electrons. Due to spin inversion symmetry, this current is exactly equal to the current carried by spin-down electrons. Therefore the total current $j \to i$ is equal to $2J_{ji}$. The current acts as a source that generates correlations s_{ij} and c_{ij}. The correlations relax with the rates $r_{s,ij}$ and $r_{c,ij}$, respectively. The relaxation is due to the hops between sites i, j, and other sites of the system. The relaxation rate of the spin correlation $r_{s,ij}$ contains the term $1/\tau_s$, which is related to the spin relaxation time and is dependent on the applied magnetic field.

To make the system of equations complete, we should write the expression for J_{ji} in terms of correlations s_{ij} and c_{ij}:

$$\begin{aligned}J_{ji} &= W_{ji}(1 - 2\overline{n}_{i\uparrow} - 2\overline{p}_{j\uparrow} + 4\overline{n}_{i\uparrow}\overline{p}_{j\uparrow} + 2c_{ij}) \\ &\quad - W_{ij}\left(\overline{n}_{i\uparrow}\overline{p}_{j\uparrow} + \frac{c_{ij} + s_{ij}}{2}\right).\end{aligned} \tag{8}$$

These equations give the general picture of the effect of correlations on the transport properties. The correlations i-j are absent in equilibrium state. They are generated by current

J_{ji} as it is shown in Eqs. (5) and (6) and have some relaxation rate related to the hopping between sites i, j and other sites of the system. The correlations s_{ij} and c_{ij} enter the expression (8) for the current indicating that the correlations can alter the transport properties of the system. Note that the correlations enter Eq. (8) in addition to the terms with averaged filling number. It means that in the nonequilibrium state of the system where all the filling numbers are equilibrium but the correlations c_{ij} and s_{ij} are nonzero, the current can flow in the system. It is important to underline that the correlations cannot be neglected even in the linear response regime. In Ref. [21], it was presumed that the correlations should influence the effective "resistances" and the transport only at high voltages.

The relaxation rate of the correlation s_{ij} is dependent on τ_s and therefore the spin relaxation time influences the transport properties even when the magnetization is zero, i.e., in the situation of the spin inversion symmetry.

These expressions correspond to the situation when site i is of type A and site j is of type B. In the opposite situation when site i is B type and site j is A type, Eqs. (5)–(8) can still be used with the substitution $i \leftrightarrow j$. For example, the spin correlation s_{ij} will be generated not with the current J_{ji} but with current $J_{ij} = -J_{ji}$. In the situation when the two sites have the same type, the expressions for the correlations are different.

Let us consider the two A-type sites i and i' [Fig. 3(a)]. The two correlations of the filling numbers on these sites can be defined as $s_{ii'} = \overline{n_{i\uparrow}n_{i'\uparrow}} - \overline{n_{i\uparrow}n_{i'\downarrow}}$ and $c_{ii'} = \overline{n_{i\uparrow}n_{i'\uparrow}} + \overline{n_{i\uparrow}n_{i'\downarrow}} - 2\overline{n}_{i\uparrow}\overline{n}_{i'\uparrow}$. It appears that the correlation $s_{ii'}$ has no source, i.e., its expression is $ds_{ii'}/dt = -r_{s,ii'}s_{ii'}$. Therefore, although this correlation can in principle exist in a nonequilibrium state, it is not generated by the applied voltage. We will consider that such correlations are absent in the system. The correlation $c_{ii'}$ is generated by the current and can influence the current:

$$\frac{dc_{ii'}}{dt} = 2(\overline{n}_{i\uparrow} - \overline{n}_{i'\uparrow})J_{i'i} - r_{c,ii'}c_{ii'},$$
$$r_{c,ii'} = \sum_{k\neq i,i'}(2W_{ki}\overline{n}_{k\uparrow} + W_{ik}\overline{p}_{k\uparrow}) + \sum_{m\neq i,i'}(2W_{mi'}\overline{n}_{m\uparrow} + W_{i'm}\overline{p}_{m\uparrow}), \quad (9)$$
$$J_{i'i} = W_{i'i}(\overline{n}_{i'\uparrow} - 2\overline{n}_{i'\uparrow}\overline{n}_{i\uparrow} - c_{ii'}) - W_{ii'}(\overline{n}_{i\uparrow} - 2\overline{n}_{i\uparrow}\overline{n}_{i'\uparrow} - c_{ii'}). \quad (10)$$

The situation in pairs of B sites is similar to the situation in pairs of A sites. In the pair j-j' of two B-type sites [Fig. 3(d)], one can consider two correlations, $s_{jj'} = \overline{p_{j\uparrow}p_{j'\uparrow}} - \overline{p_{j\uparrow}p_{j'\downarrow}}$ and $c_{jj'} = \overline{p_{j\uparrow}p_{j'\uparrow}} + \overline{p_{j\uparrow}p_{j'\downarrow}} - 2\overline{p}_{j\uparrow}\overline{p}_{j'\uparrow}$. However, the correlation $s_{jj'}$ is not generated by the current and does not contribute to the transport properties. The correlation $c_{jj'}$ is generated by the current and can influence the transport properties,

$$\frac{dc_{jj'}}{dt} = 2(\overline{p}_{j'\uparrow} - \overline{p}_{j\uparrow})J_{j'j} - r_{c,jj'}c_{jj'},$$
$$r_{c,jj'} = \sum_{k\neq j,j'}(W_{kj}\overline{n}_{k\uparrow} + 2W_{jk}\overline{p}_{k\uparrow}) + \sum_{m\neq j,j'}(W_{mj'}\overline{n}_{m\uparrow} + 2W_{j'm}\overline{p}_{m\uparrow}), \quad (11)$$
$$J_{j'j} = W_{j'j}(\overline{p}_{j\uparrow} - 2\overline{p}_{j\uparrow}\overline{p}_{j'\uparrow} - c_{jj'}) - W_{jj'}(\overline{p}_{j'\uparrow} - 2\overline{p}_{j\uparrow}\overline{p}_{j'\uparrow} - c_{jj'}). \quad (12)$$

Equations (5)–(12) are the kinetic equations for the case of infinite Hubbard energy (the model of A- and B-type sites). In this case, each pair of sites i and j can be described with four values: $\overline{n}_{i\uparrow}$, $\overline{n}_{j\uparrow}$, the spin correlation s_{ij}, and the charge correlation c_{ij}. Similar considerations are possible also for the general case of arbitrary Hubbard energy. In this case, each site can have zero, one, or two electrons, and can be described by two independent filling numbers $\overline{n}_{i\uparrow}$ and $\overline{n}_{i2}$—the probability for site i to have one electron with spin up and two electrons, respectively. The number of independent correlations ν_{ij} between sites i and j is equal to five. We describe them with the vector $\vec{\nu}_{ij}$:

$$\vec{\nu}_{ij} = \begin{pmatrix} s_{ij} \\ c_{ij} \\ \nu_{ij,2\uparrow} \\ \nu_{ij,\uparrow 2} \\ \nu_{ij,22} \end{pmatrix} = \begin{pmatrix} s_{ij} \\ c_{ij} \\ \overline{n_{i2}n_{j\uparrow}} - \overline{n}_{i2}\overline{n}_{j\uparrow} \\ \overline{n_{i\uparrow}n_{j2}} - \overline{n}_{i\uparrow}\overline{n}_{j2} \\ \overline{n_{i2}n_{j2}} - \overline{n}_{i2}\overline{n}_{j2} \end{pmatrix}. \quad (13)$$

The additional correlations are related to the possibility for each site to play both roles: of A site and of B site. These additional correlations become proportional to c_{ij} in the limit $E_{\rm hub} \to \infty$. Only one correlation s_{ij} is directly related to the spin relaxation rate. Therefore the new correlations can be considered as additional charge correlations.

In this general case, it is useful to introduce four currents flowing between sites i and j, J_{ij}^X, where indices X describe the role played by the sites and can have one of four values AA, AB, BA, or BB. For example, the current J_{ij}^{AB} stands for the hop of the first electron on site i to the single-occupied site j and to the backward hop of second electron from site j to the empty site i. All four currents are equal to zero in the equilibrium.

The general picture of correlations affecting the electron kinetics is similar to the one in the model of A-type and B-type sites. The correlations ν_{ij}^α are generated by the currents J_{ji}^X and relax due to hops to other sites and the spin relaxation:

$$\frac{d}{dt}\nu_{ij}^\alpha = G_{ij}^{\alpha X}J_{ji}^X - R_{ij}^{\alpha\beta}\nu_{ij}^\beta. \quad (14)$$

Here, indices α and β stand for the components of vector $\vec{\nu}_{ij}$. The correlations $\vec{\nu}_{ij}$ contribute to the currents in an additive way:

$$J_{ji}^X = J_{ji,0}^X + \mathcal{W}_{ij}^{X\alpha}\nu_{ij}^\alpha. \quad (15)$$

Here, $J_{ji,0}^X$ are the expressions for currents that neglect correlations. $G_{ij}^{\alpha X}$, $R_{ij}^{\alpha\beta}$, and $\mathcal{W}_{ij}^{X\alpha}$ are the matrices that describe the generation of correlations by currents, relaxation of correlations, and the effect of correlations on currents correspondingly. The explicit form of these matrices is rather cumbersome and we present them in Ref. [22] along with the explicit expression for $J_{ji,0}^X$.

IV. RESISTOR NETWORK

The kinetic equation for the hopping transport can be linearized in the limit $eEr_{ij} \ll k_BT$, where E is the applied electric field and r_{ij} is the distance between hopping sites. In the conventional hopping theory, this linearization yields the Miller-Abrahams resistor network. In the present section, we show that the linearization of our equations (5)–(12) for the model of A- and B-type sites leads to a generalized resistor network where the conductivities of resistors connecting A and B sites explicitly depend on the spin relaxation time τ_s. Let us note that the resistor network approach was applied for the description of OMAR in Ref. [28], where a node of the network was related to the many-body states of the system. This approach is different from our generalized Miller-Abrahams network where the nodes of the network correspond to the localization sites.

In the linearized equation, the relaxation rates $r_{s,ij}$ and $r_{c,ij}$ should be calculated in the equilibrium. Therefore the equilibrium values of $\overline{n}_{k\uparrow}$ and $\overline{p}_{k\uparrow}$ should be substituted in (5) and (7). The relaxation rates $r_{s,ij}$ and $r_{c,ij}$ then appear to have constant values determined by the configuration of the disorder.

Equations (5) and (6) can be reduced to a matrix equation for the correlators

$$\begin{pmatrix} r_{s,ij}+\frac{W_{ij}}{2} & \frac{W_{ij}}{2}-2W_{ji} \\ \frac{W_{ij}}{2} & \widetilde{r}_{c,ij}+\frac{W_{ij}}{2}-2W_{ji} \end{pmatrix}\begin{pmatrix} s_{ij} \\ c_{ij} \end{pmatrix}=\begin{pmatrix} 1 \\ 1 \end{pmatrix}J_{ji}^{(0)},$$
$$\widetilde{r}_{c,ij}=r_{c,ij}/(1-2\overline{n}_{i\uparrow}-2\overline{p}_{j\uparrow}). \tag{16}$$

Here, $J_{ji}^{(0)}$ is the term in the current that does not include the correlators. It can be expressed in terms of Miller-Abrahams resistance $R_{ij}^{(MA)}$ of the pair i-j and the voltage u_{ji} applied to the pair $J_{ji}^{(0)}=u_{ji}/R_{ij}^{(MA)}$. $\widetilde{r}_{c,ij}=r_{c,ij}/(1-2\overline{n}_{i\uparrow}-2\overline{p}_{j\uparrow})$.

Equation (16) should be solved and the values of the correlators c_{ij} and s_{ij} should be substituted into Eq. (8). This leads to the following equation for the resistor that includes the effect of pair intersite correlations:

$$R_{ij}^{(AB)}=R_{ij}^{(MA)}\left(1+\frac{W_{ij}}{(2/\tau_s)+2r_{s,ij}^{(0)}}+\frac{W_{ij}-4W_{ji}}{2\widetilde{r}_{c,ij}}\right). \tag{17}$$

Equation (17) demonstrates that the correlations enter the expression for the resistor as an additional multiplier. This multiplier contains the term $W_{ij}/(2/\tau_s+2r_{s,ij}^{(0)})=W_{ij}/2r_{s,ij}$, which depends on the applied magnetic field. It is also clear from Eq. (17) that the discussed mechanism of OMAR leads to the positive magnetoresistance. Naturally, $r_{s,ij}$ decreases with applied magnetic field leading to the increase in the resistance $R_{ij}^{(AB)}$ because the term $W_{ij}/2r_{s,ij}$ is always positive. The second term $(W_{ij}-4W_{ji})/2\widetilde{r}_{c,ij}$ is related to charge correlations c_{ij}. It can have arbitrary sign but does not depend on τ_s and on the applied magnetic field.

The resistors that connect pairs of sites of equal type can be treated in the same way. The corresponding expressions for resistances $R_{ii'}^{(AA)}$ and $R_{jj'}^{(BB)}$ are

$$R_{ii'}^{(AA)}=R_{ii'}^{(MA)}\left(1+\frac{2(\overline{n}_{i\uparrow}-\overline{n}_{i'\uparrow})(W_{i'i}-W_{ii'})}{r_{c,ii'}}\right), \tag{18}$$

$$R_{jj'}^{(BB)}=R_{jj'}^{(MA)}\left(1+\frac{2(\overline{p}_{j\uparrow}-\overline{p}_{j'\uparrow})(W_{jj'}-W_{j'j})}{r_{c,jj'}}\right). \tag{19}$$

The resistances $R_{ii'}^{(AA)}$ and $R_{jj'}^{(BB)}$ are dependent on the charge correlation $c_{ii'}$ but not on spin correlation $s_{ii'}$. Therefore the magnetic field does not enter the expression for these resistances.

Equations (17)–(19) reduce the problem of A- and B-type sites at low applied voltage to the network of classical resistors. This network can be treated by the same method as a classical Miller-Abrahams resistor network, for example, with percolation theory or with direct numerical solution of the Kirchhoff equations, which is much easier than a Monte Carlo simulation.

The linearization of the kinetic equations in the case of arbitrary Hubbard energy is discussed in Supplemental Material [22]. In the general case, the linearization yields an analog of the Kirchhoff equations—a system of linear equations that can be solved to find the site potentials and the currents. However, the linear equations in this case cannot be reduced to an equivalent scheme that contains only resistances.

V. NUMERICAL SIMULATION

The kinetic equations (14) and (15) describe the microscopic response of pairs of sites to the applied electric field. The calculation of the magnetoresistance in a macroscopic sample requires an averaging over the sample. In principle, the linearized version of kinetic equations allows us to apply conventional methods of averaging such as the percolation theory in a resistance network with an exponentially broad distribution of resistances [4], but this is beyond the scope of the present study.

We follow an alternative way and solve the kinetic equations numerically. We consider the general case, i.e., do not restrict our simulations to the model of A- and B-type sites and to small applied electric fields. The main goal of the present simulation is to show the most general features of the magnetoresistance described with the kinetic equations (14) and (15) and compare them with the other existing models, such as the percolation theory based on the momentary filling numbers [14–16].

In our simulations, we apply the standard Euler method. Similar to Ref. [29], we apply periodic boundary conditions in the presence of an external electric field. For numerical simulations, we use a square lattice with the size 32×32 and 64×64 sites with energetic disorder $-\Delta E/2\leqslant E_{i,j}\leqslant\Delta E/2$ and with Miller-Abrahams nearest-neighbor hopping rates $W_{i,j}=\omega_0\exp\left(-[\Delta E_{i,j}+|\Delta E_{i,j}|]/2k_BT\right)$, where ω_0 is a prefactor and $\Delta E_{i,j}$ is the energy difference between sites j and i including the contribution from the external electric field and Hubbard energy E_{hub} in the case of double occupation. The logarithms of the conductances [29] are averaged over 100 random energy configurations. To make sure that the size effects are small, we have compared the calculated magnetoresistance for the systems 32×32 and 64×64 sites. As it follows from the comparison presented in Ref. [22], the finite size effects are negligible. Similarly, we have compared the results of magnetoresistance calculations for different number of averaging over random energy configurations. The results presented in Ref. [22] suggest that averaging over 100 random energy configurations provides reliable results for the considered parameters of the system. The results of

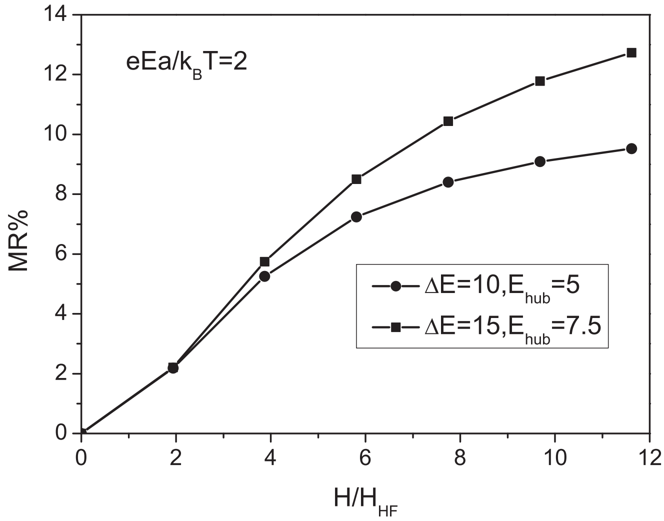

FIG. 4. The dependence of the magnetoresistance on magnetic field. Here we assume that the spin relaxation rate is determined by Eq. (1) with $\omega_s^2/\omega_0^2 = 1/15$.

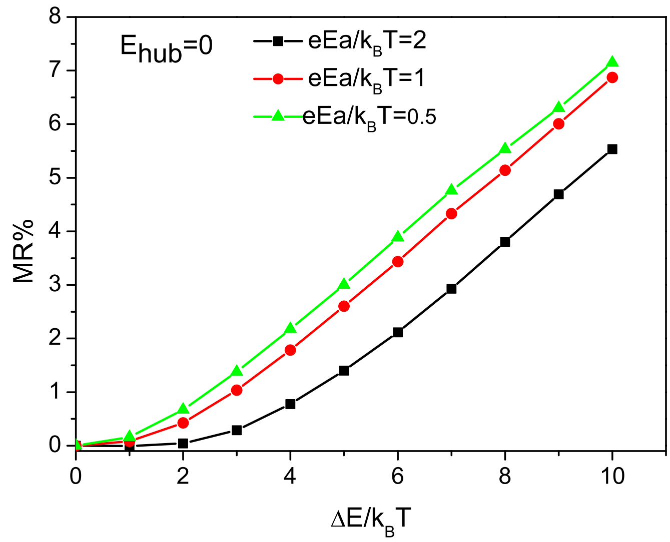

FIG. 5. The dependence of the magnetoresistance on disorder for different electric fields.

calculations of the magnetoresistance are presented in Fig. 4. The magnetoresistance quickly increases at small magnetic field $H \leqslant H_{\rm HF}$ and then slowly approaches its limiting value $MR \approx MR_\infty - {\rm const} H^{-2}$ at $H \gg H_{\rm HF}$. Note that in the presented case the limiting magnetoresistance is about $MR_\infty \approx$ 10% but it is increasing with the increase of disorder and Hubbard energy.

It is interesting that the magnetoresistance is absent for the absence of disorder $\Delta E = 0$ and zero Hubbard energy $E_{\rm hub} = 0$. It can be shown analytically that in this case the equilibrium filling numbers and zero intersite correlations solve the kinetic equations in arbitrary applied field. Therefore no organic magnetoresistance can be observed in such a system. Indeed, assuming that n_{i2} and $n_{i\uparrow}$ are equilibrium filling numbers and taking into account that $n_{i2}(1 - 2n_{i\uparrow} - n_{i2}) = n_{i\uparrow}^2$, we can write the expression for uniform currents: $J_{ij}^{AA} = \omega_0 n_{i\uparrow}(1 - 2n_{i\uparrow} - n_{i2})(1 - \exp(-eEa/k_BT))$, $J_{ij}^{AB} = J_{ij}^{BA} = \omega_0 n_{i\uparrow}^2[1 - \exp(-eEa/k_BT)]$, $J_{ij}^{BB} = \omega_0 n_{i\uparrow} n_{i2} (1 - \exp(-eEa/k_BT))$. Substituting these currents into Eq. (14), it is easy to see that the nonuniform part of this equation containing currents reduces to zero. It means that all the correlations go to zero, $\nu_{i,j}^\alpha = 0$. On the other hand, the charge conservation equations (Eqs. (22) and (23) from Ref. [22]) are satisfied automatically for uniform currents. Physically, it means that although the hopping rates are modified by the applied electric field and the current flows, the distribution of charges and spins remains at equilibrium. The spin relaxation has no effect on the equilibrium distribution of spins and does not influence the charge transport. Note, however, that this is the case only for relatively simple ordered systems like the ordered square lattice. In a system with two or more site types with different energies forming some sort of a complex lattice, the organic magnetoresistance should be nonzero. To the best of our knowledge, the absence of OMAR in these conditions was never reported and it was not predicted by models [14–16].

The increase of disorder in the absence of $E_{\rm hub} = 0$ leads to the increase of magnetoresistance, Fig. 5. Similarly, magnetoresistance appears when the disorder is absent, $\Delta E = 0$, and $E_{\rm hub}$ is finite, Fig. 6. Note that the magnetoresistance has a characteristic maximum when $E_{\rm hub} \approx eEa/k_BT$. Further increase of Hubbard energy leads to a decrease of magnetoresistance. These results reflect the absence of correlations in an ordered system with zero Hubbard energy. The increase of disorder and the finite Hubbard energy lead to the appearance of nonzero correlations and to finite magnetoresistance.

The predicted magnetoresistance is finite in the limit of the weak field, $eEa/k_BT \ll 1$, and has relatively weak dependence on the external electric field. It increases by 30% in the limit $eEa/k_BT \gg 1$ for $E_{\rm hub} > eEa$ and decreases by 30% for the case $E_{\rm hub} < eEa$, see Fig. 7. The increase of the

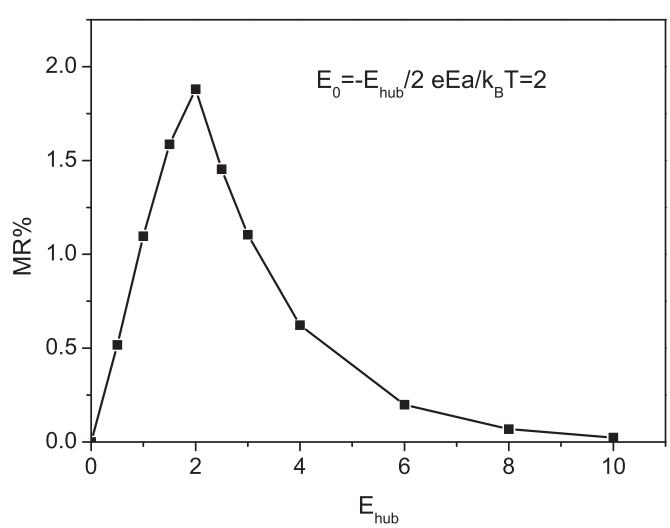

FIG. 6. The dependence of the magnetoresistance on the Hubbard energy. All the sites are considered to have the same single-occupation energy, $E_0 = -E_{\rm hub}/2$.

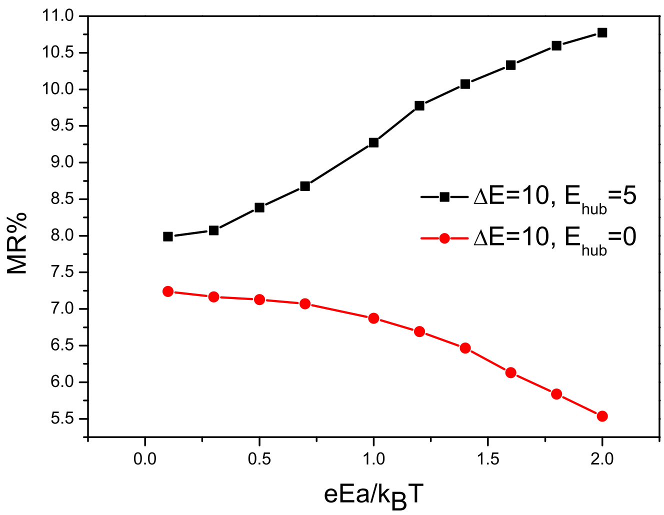

FIG. 7. The dependence of the magnetoresistance on the external electric field.

magnetoresistance in the limit of finite E_{hub} with increasing electric field agrees with the results obtained in Ref. [7].

VI. DISCUSSION

Some of the previous attempts to include OMAR into the analytical theory of the hopping transport considered the percolation theory with momentary spin projections, which effectively corresponds to the percolation with momentary filling numbers. For example, the number of sites available for the hop was counted as the number of free sites and the sites with one electron with the spin antiparallel to the spin of the hopping electron. While such a scheme leads to a finite magnetoresistance, it is inconsistent with the general rules that are used in the well-known problem of hopping transport. To clarify that, let us consider the simple problem of neighbor hopping over sites with random positions without double occupation possibility and assume that the number of electrons is half the number of sites. The idea of the percolation theory with the momentary filling numbers means that only half of sites are available for the hop because the other half is filled. However, in the percolation theory for this problem [4], all the sites are counted because a filled site close to the initial site of the hop will typically lose its electron faster than the time of the hop to a more distant site (that is exponentially larger than the time of the hop to the neighbor site). The previous approaches [14–16] do not consider the possibility for the target site of the hop (for example, with one electron with the parallel spin configuration) to lose its electron due to some other hop. Note that previous attempts to make an analytical theory of bipolaron mechanism of OMAR did not link this magnetoresistance with the intersite correlations of the filling numbers. Here we show that the relation is quite direct. The absence of correlations, for example, in an ordered system with zero Hubbard energy, means absence of magnetoresistance.

We propose another approach to include OMAR into the analytic theory of hopping transport by including the correlations in the kinetic equations. The shape of the dependence of resistivity on the applied magnetic field in our theory is similar to the one in previous theories. It is governed by the dependence $\tau_s(H)$, which is not closely related to the physics of hopping conduction. However, the dependence of the magnetoresistance on system parameters is different. The most clear example of this difference is the absence of magnetoresistance in an ordered system with zero Hubbard energy.

In our theory, we considered only pair intersite correlations neglecting the triple correlations. We also neglected "long-range" correlations between sites that are not "connected" with effective hopping. The main reason for these assumptions is that the kinetic equations with pair correlations is the minimal model that allows us to include OMAR into a conventional theory of the hopping transport. The neglected correlations can, in principle, modify the organic magnetoresistance but cannot completely suppress it. Note that neglecting triple correlations is a controllable approximation when the number of electrons is small and the number of B-type sites is small (in the model of A-type and B-type sites). In this case, the problem allows the expansion into series over the correlation order. Organic magnetoresistance appears in the second order of the expansion (pair correlations). The higher order correlations can lead only to a small perturbation in this situation.

Another assumption made in our theory is the simplified treatment of the spin dynamics that was reduced to a single spin relaxation time. To consider the effect of realistic spin rotation in the hyperfine field on the conductivity, one should include this rotation into the kinetic equations for the correlations. However, it will not only influence the dynamics of the discussed spin correlation s_{ij}, but also will lead to the appearance of new spin correlations. It is similar to the appearance of new charge correlations due to finite Hubbard energy. In the general case, the spin state of two electrons can be described by a 4×4 density matrix. However, in our case, the system is invariant under rotations of the spin space. It significantly reduces the possible form of the density matrix. The only nonequilibrium form of the matrix in the stationary case is related to the possible imbalance between triplet and singlet electron pairs. It can be expressed in terms of a single spin correlation. However, if we introduce real hyperfine fields on the localization sites, these fields will introduce a preferred spin direction. New spin correlations that reflect the appearance of average values similar to $\langle[(\mathbf{S}_i + \mathbf{S}_j)(\mathbf{H}_i - \mathbf{H}_j)]^2\rangle$ are important for the problem. Therefore, to make a correlation-based theory of OMAR, one should identify the spin correlations that appear in the general case and discuss their dynamics in the presence of the local hyperfine fields, the spin-orbit coupling, and the exchange interaction.

The absence of the bipolaron OMAR in an ordered system with zero Hubbard energy is not sensitive to our treatment of the higher-order correlations. Naturally, correlations of different order can be connected with the Bogoliubov-Born-Green-Kirkwood-Yvon chain of equations [30]. This chain of equations shows that correlations of order $n+1$ can be generated by correlations of order n. We have shown that pair correlations are not generated in the ordered system with $E_{\text{hub}} = 0$. Triple correlations will not be generated because of the absence of pair correlations to generate them. The same applies to the realistic spin dynamics. This dynamics should not influence the density matrix that describes the equilibrium distribution of spins and charges. Before the dynamics of the spin correlations becomes important, these correlations should be generated. Note that the only spin correlation that can be generated due to the hopping is the difference between the singlet and triplet probabilities proportional to s_{ij}. In the ordered system with $E_{\rm hub} = 0$, spin correlations are not generated and their spin dynamics is not essential.

The bipolaron mechanism discussed in the present study is the most probable mechanism of organic magnetoresistance in materials with a single carrier type. However, in many experiments, OMAR was measured in bipolar devices where the current is carried by both electrons and holes. The physics of such devices is more complex. The effect of the magnetic field on the current can be related to the formation and dissociation of excitons [5,6,31] or to the interaction of excitons with charge carriers [32,33]. In many cases, it is still attributed to the effect of magnetic field on the rate of the hyperfine spin relaxation. The difference from the bipolaron mechanism is related to the fact that the mutual spin orientation of the charge carriers influences not the process of bipolaron formation but other processes such as the formation of excitons from electron-hole pairs. We believe that the method proposed in the present study can be generalized to account for this mechanism. However, in this case, both electron and hole filling numbers should be included into the kinetic equations along with the corresponding correlations between filling numbers of electrons and holes.

In conclusion, we derive kinetic equations for the hopping conduction with a double occupation possibility that include pair correlations. Contrary to the conventional equations that are based on Hartree decoupling, our equations reflect the dependence of the conductivity on the spin relaxation time even without net polarization and can describe OMAR. In the linear regime, the equations can be reduced to the generalized resistor network. OMAR described with our equations have a dependence on the magnetic field similar to the previous studies but have a different dependence on system parameters.

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