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Theory of quantum diffusion in biased semiconductors

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Abstract

A general theory is developed to describe diffusion phenomena in biased semiconductors and semiconductor superlattices. It is shown that the Einstein relation is not applicable for all field strengths so that the calculation of the field-mediated diffusion coefficient represents a separate task. Two quite different diffusion contributions are identified. The first one disappears when the dipole operator commutes with the Hamiltonian. It plays an essential role in the theory of small polarons. The second contribution is obtained from a quantity that is the solution of a kinetic equation but that cannot be identified with the carrier distribution function. This is in contrast to the drift velocity, which is closely related to the distribution function. A general expression is derived for the quantum diffusion regime, which allows a clear physical interpretation within the hopping picture.

1. Introduction

Carrier motion in quantum systems exhibits a variety of different behaviours including ballistic transport, diffusion and localization. Each of these features of carrier motion has attracted a great deal of interest. The quantum theory of high-field carrier transport has been developed to a high level of sophistication. In contrast, there does not seem to be much theoretical work addressing the detailed microscopic description of diffusion processes in semiconductors under the application of electric fields of arbitrary strength. This will be the subject of this paper, in which we focus on a general treatment of electric-field driven carrier diffusion in semiconductors.

The most interesting aspect that we are confronted with in a study of diffusion phenomena is quantum diffusion, when particles tunnel through energy barriers. An example of quantum diffusion is the motion of muonium atoms in solids [1]. There are different modes of quantum diffusion depending on the type of interaction with host atoms, including the competition between localization and delocalization. As muonium atoms are heavier than electrons, quantum diffusion applies more than ever to electrons, which are localized by random potentials

or strong electric fields. Moreover, the concept of quantum diffusion has been applied to completely different areas of physics, for instance to the treatment of open quantum systems within the quantum state diffusion picture [2] or to numerical calculations based on the diffusion Monte Carlo approach [3].

Based on semiclassical Boltzmann or balance equations as well as the nonequilibrium Green function technique, the theory of electron transport in semiconductors has advanced considerably in recent years. Contrary to this achievement, there are only a few microscopic treatments of the field-induced carrier diffusion in semiconductors. Considering the quantum Brownian motion of a single electron in a superlattice (SL), the electron diffusion coefficient has been calculated within a one-dimensional model [4]. The results apply to the Wannier–Stark (WS) transport regime, where the electron drift velocity is due to inelastic scattering. Taking into account scattering on acoustic phonons, Mourokh *et al* [4] obtained the result that the electron diffusion vanishes at the positions of the electron–phonon resonances. The derived analytical expressions for the drift velocity and the diffusion coefficient have been used for a microscopic analysis of field-domain formation in SLs [5]. This work complements a former approach by Laikhtman and Miller [6], who presented a detailed microscopic theory of current–voltage instabilities in SLs. However, to our knowledge there is no general approach to carrier diffusion in semiconductors, which covers all electric-field strengths ranging from the ohmic to the WS transport regime. It is the aim of this paper to fill this gap by presenting a systematic microscopic theory of field-mediated carrier diffusion in semiconductors and semiconductor SLs.

2. Basic theory

In this section, a microscopic theory is presented from which general expressions for the field-dependent average drift velocity and diffusion coefficient are derived. The approach is restricted to the single-particle picture of a nondegenerate electron gas, which occupies only one energy band. The treatment of interband (or intersubband) transitions as well as the Coulomb interaction will be the subject of further work. We consider the situation when, after the electric field is switched on, the electron distribution gradually approaches a homogeneous state. Most interesting is the behaviour of the system at sufficiently long times after switching on the field ($t \to \infty$). The consideration of this limit is carried out in Laplace space ($s \to 0$). The quantity of most interest is the diffusion propagator P, which satisfies the following phenomenological diffusion equation formulated in Laplace space

$$sP(\boldsymbol{r}-\boldsymbol{r}_0|s) = \delta(\boldsymbol{r}-\boldsymbol{r}_0) + v_z(s)\frac{\partial}{\partial z}P(\boldsymbol{r}-\boldsymbol{r}_0|s) + D_{zz}(s)\frac{\partial^2}{\partial z^2}P(\boldsymbol{r}-\boldsymbol{r}_0|s). \tag{1}$$

Here, $v_z(s)$ and $D_{zz}(s)$ denote the average drift velocity along the SL axis and a component of the diffusion tensor respectively. These quantities are calculated from the moments of the diffusion propagator

$$Z_n(s) = \int \mathrm{d}^3\boldsymbol{r}\,(z-z_0)^n P(\boldsymbol{r}-\boldsymbol{r}_0|s). \tag{2}$$

Carrying out the calculations, we obtain

$$v_z(s) = -s^2 Z_1(s), \tag{3}$$

$$D_{zz}(s) = \frac{s^2}{2}Z_2(s) - \frac{v_z(s)^2}{s}, \tag{4}$$

where $Z_0(s) = 1/s$ as derived from equation (1) has been taken into account. From the point of view of a microscopic approach, the diffusion propagator $P(\boldsymbol{r}-\boldsymbol{r}', t)$ has to be identified

with the conditional probability of finding a particle near the point $\boldsymbol{r}'$ at time t, provided it occupied the lattice site $\boldsymbol{r}$ at an earlier time $t = 0$. Using the Wannier representation, this probability is calculated by an inverse Laplace transformation from the diagonal components of the following vacuum expectation value [7]

$$P^{m_1 m_3}_{m_2 m_4}(s) = \frac{1}{Z}\int_0^\infty \mathrm{d}t\, \mathrm{e}^{-st}\,\mathrm{Sp}_{ph}\left\{\mathrm{e}^{-H_{ph}/k_BT}\langle 0|a_{m_2}\mathrm{e}^{\mathrm{i}Ht/\hbar}a^+_{m_4}a_{m_3}\mathrm{e}^{-\mathrm{i}Ht/\hbar}a^+_{m_1}|0\rangle\right\}, \tag{5}$$

averaged over the vibrational subsystem described by the Hamiltonian H_{ph}. From equation (5), we deduce the quantity $P(\boldsymbol{r} - \boldsymbol{r}_0|s) \equiv P^{rr_0}_{rr_0}(s)$, which is used to calculate the moments in equation (2). The fermionic creation and annihilation operators are denoted by a^+_m and a_m. The averaging is carried out over the vibrational degrees of freedom. The partition function is expressed by $Z = \mathrm{Tr}\{\exp(-H_{ph}/k_BT)\}$. The total Hamiltonian

$$H = H' + H_E, \quad \text{with} \quad H' = H_e + H_{ph} + H_{int} \tag{6}$$

of our system contains the contribution of the electric field

$$H_{\mathcal{E}} = -e\mathcal{E}\cdot\sum_m a^+_m \boldsymbol{R}_m a_m, \tag{7}$$

the free electron part

$$H_e = \sum_{m,m'} \varepsilon(\boldsymbol{m} - \boldsymbol{m}')a^+_m a_{m'}, \tag{8}$$

with the dispersion relation $\varepsilon(\boldsymbol{k})$ in the Fourier space, and the interaction term H_{int}, which accounts for elastic scattering on lattice defects and inelastic scattering on phonons. The treatment of the diffusion propagator P proceeds by a diagrammatic analysis, from which a Bethe–Salpeter equation is derived [8]

$$P^{m_1 m_3}_{m_2 m_4}(s) = R^{m_1 m_3}_{m_2 m_4}(s) + \sum_{\{m'_i\}} P^{m_1 m'_1}_{m_2 m'_2}(s) W^{m'_1 m'_3}_{m'_2 m'_4}(s) R^{m'_3 m_3}_{m'_4 m_4}(s), \tag{9}$$

with W being the scattering probability calculated from the interaction part H_{int} of the Hamiltonian (in our general approach the form of H_{int} is not specified). R denotes the diffusion propagator for a system of independent particles ($H_{int} = 0$), which is expressed by

$$R^{m_1 m_3}_{m_2 m_4}(s) = \int_0^\infty \mathrm{d}t\, \mathrm{e}^{-st}\langle 0|a_{m_2}\mathrm{e}^{\mathrm{i}(H_e+H_E)t/\hbar}a^+_{m_4}a_{m_3}\mathrm{e}^{-\mathrm{i}(H_e+H_E)t/\hbar}a^+_{m_1}|0\rangle. \tag{10}$$

An equation of motion for this quantity is derived by integration by parts. The calculations are most suitably carried out in the symmetry-adapted Wigner representation of the correlation functions [8]

$$P^{m_1 m_3}_{m_2 m_4}(s) = \frac{1}{N^2}\sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{\kappa}} P(\boldsymbol{k},\boldsymbol{k}',\boldsymbol{\kappa}|s)\exp\Bigg\{\{\mathrm{i}\boldsymbol{k}(\boldsymbol{R}_{m_2} - \boldsymbol{R}_{m_1}) - \mathrm{i}\boldsymbol{k}'(\boldsymbol{R}_{m_4} - \boldsymbol{R}_{m_3}) + \frac{\mathrm{i}}{2}\boldsymbol{\kappa}(\boldsymbol{R}_{m_1} + \boldsymbol{R}_{m_2} - \boldsymbol{R}_{m_3} - \boldsymbol{R}_{m_4})\}\Bigg\}. \tag{11}$$

Using this representation, the equation of motion of the bare diffusion propagator R has the form

$$[s + \hat{I}(\boldsymbol{k}',\boldsymbol{\kappa})]R(\boldsymbol{k},\boldsymbol{k}',\boldsymbol{\kappa}|s) = \delta_{\boldsymbol{k},\boldsymbol{k}'}, \tag{12}$$

where the differential operator

$$\hat{I}(\boldsymbol{k}',\boldsymbol{\kappa}) = \frac{e\mathcal{E}}{\hbar}\nabla_{\boldsymbol{k}'} - \frac{\mathrm{i}}{\hbar}\left[\varepsilon\left(\boldsymbol{k}' + \frac{\boldsymbol{\kappa}}{2}\right) - \varepsilon\left(\boldsymbol{k}' - \frac{\boldsymbol{\kappa}}{2}\right)\right] \tag{13}$$

has been introduced. Applying this operator on both sides of the Wigner transformed version of equation (9), we obtain

$$[s + \hat{I}(\boldsymbol{k}', \boldsymbol{\kappa})]P(\boldsymbol{k}, \boldsymbol{k}', \boldsymbol{\kappa}|s) = \delta_{\boldsymbol{k},\boldsymbol{k}'} + \sum_{\boldsymbol{k}_1} P(\boldsymbol{k}, \boldsymbol{k}_1, \boldsymbol{\kappa}|s)W(\boldsymbol{k}_1, \boldsymbol{k}', \boldsymbol{\kappa}|s). \tag{14}$$

This equation has to be solved under the restriction that the sum rule

$$\sum_{\boldsymbol{k}'} P(\boldsymbol{k}, \boldsymbol{k}', \boldsymbol{\kappa} = \boldsymbol{0}|s) = \frac{1}{s} \tag{15}$$

is satisfied [8]. This sum rule follows from equation (14) by taking into account the relation of detailed balance $\sum_{\boldsymbol{k}'} W(\boldsymbol{k}, \boldsymbol{k}', \boldsymbol{\kappa} = 0|s) = 0$. Equation (14) together with the condition (15) represents our basic results, which allows a straightforward calculation of the moments

$$Z_n(s) = \mathrm{i}^n \sum_{\boldsymbol{k},\boldsymbol{k}'} \frac{\partial^n}{\partial \kappa_z^n} P(\boldsymbol{k}, \boldsymbol{k}', \boldsymbol{\kappa}|s)|_{\boldsymbol{\kappa}=\boldsymbol{0}} \equiv \mathrm{i}^n \sum_{\boldsymbol{k},\boldsymbol{k}'} P_n(\boldsymbol{k}, \boldsymbol{k}'|s). \tag{16}$$

From these quantities, the drift velocity and the diffusion coefficient are easily obtained. The correlation functions P_1 and P_2, which enter equation (16), are determined from the kinetic equation (14). The formal solutions are expressed by

$$P_1(\boldsymbol{k}, \boldsymbol{k}'|s) = \sum_{\boldsymbol{k}_1} P(\boldsymbol{k}_1, \boldsymbol{k}'|s)\left\{\mathrm{i}v_z(\boldsymbol{k}_1)P(\boldsymbol{k}, \boldsymbol{k}_1|s) + \sum_{\boldsymbol{k}_2} P(\boldsymbol{k}, \boldsymbol{k}_2|s)W_1(\boldsymbol{k}_2, \boldsymbol{k}_1|s)\right\}, \tag{17}$$

$$P_2(\boldsymbol{k}, \boldsymbol{k}'|s) = \sum_{\boldsymbol{k}_1} P(\boldsymbol{k}_1, \boldsymbol{k}'|s)\left\{2\mathrm{i}v_z(\boldsymbol{k}_1)P_1(\boldsymbol{k}, \boldsymbol{k}_1|s)\right.$$
$$\left. + 2\sum_{\boldsymbol{k}_2} P_1(\boldsymbol{k}, \boldsymbol{k}_2|s)W_1(\boldsymbol{k}_2, \boldsymbol{k}_1|s) + \sum_{\boldsymbol{k}_2} P(\boldsymbol{k}, \boldsymbol{k}_2|s)W_2(\boldsymbol{k}_2, \boldsymbol{k}_1|s)\right\}, \tag{18}$$

where the abbreviations $W_n(\boldsymbol{k}, \boldsymbol{k}'|s) = \partial^n W(\boldsymbol{k}, \boldsymbol{k}', \boldsymbol{\kappa}|s)/\partial\kappa_z^n|_{\boldsymbol{\kappa}=0}$ and $v_z(\boldsymbol{k}) = (1/\hbar)\partial\varepsilon(\boldsymbol{k})/\partial k_z$ have been used. The calculation of the moments Z_1 and Z_2 as well as the respective observable $v_z(s)$ and $D_{zz}(s)$ is facilitated by introducing the carrier distribution function

$$f(\boldsymbol{k}, s) = s\sum_{\boldsymbol{k}'} P(\boldsymbol{k}', \boldsymbol{k}|s). \tag{19}$$

According to the sum rule (15), the function defined in equation (19) is normalized $[\sum_{\boldsymbol{k}} f(\boldsymbol{k}, s) = 1]$. A kinetic equation for the distribution function is easily obtained from equation (14) and the definition in equation (19)

$$\left[s + \frac{e\mathcal{E}}{\hbar}\nabla_{\boldsymbol{k}}\right] f(\boldsymbol{k}, s) = s + \sum_{\boldsymbol{k}'} f(\boldsymbol{k}', s)W(\boldsymbol{k}', \boldsymbol{k}|s). \tag{20}$$

In the relevant long-time limit ($s \to 0$) and without taking into account the influence of the electric field on the scattering probability W, we recover the Boltzmann equation from equation (20). This is an additional justification for the identification of $f(\boldsymbol{k})$ with the carrier distribution function. The average drift velocity is obtained by inserting the formal solution of equation (17) into (3) and by considering the sum rule (15). The final result for the average drift velocity

$$v_z(s) = \sum_{\boldsymbol{k}} v_{eff}(\boldsymbol{k}, s)f(\boldsymbol{k}, s) \tag{21}$$

is expressed by an effective velocity [8]

$$v_{eff}(\boldsymbol{k}, s) = v_z(\boldsymbol{k}) - \mathrm{i}\sum_{\boldsymbol{k}'} W_1(\boldsymbol{k}, \boldsymbol{k}'|s), \tag{22}$$

which contains, in addition to the expected group velocity $v_z(\boldsymbol{k})$, a scattering-induced contribution associated with W_1. The latter term vanishes in most cases, when H_{int} commutes with the dipole operator. However, in the theory of small polarons or for systems with discrete energy levels, it is just this scattering-induced W_1 contribution which dominates the average drift velocity.

So far we have reproduced well the established results concerning the drift velocity that were derived and discussed many years ago [8]. In the following, the calculational scheme outlined above is applied to construct a general theory of field-mediated quantum diffusion in semiconductors. Let us proceed by exploiting the formal solution of equation (18) to calculate the second moment Z_2. Introducing the distribution function $f(\boldsymbol{k}, s)$ from equation (19) and the effective velocity from equation (22), we obtain

$$sZ_2(s) = -2\mathrm{i}\sum_{\boldsymbol{k}} v_{eff}(\boldsymbol{k}, s)P_1(\boldsymbol{k}, s) - \frac{1}{s}\sum_{\boldsymbol{k}_1,\boldsymbol{k}_2} f(\boldsymbol{k}_1, s)W_2(\boldsymbol{k}_1, \boldsymbol{k}_2|s), \tag{23}$$

where the function $P_1(\boldsymbol{k}, s) = \sum_{\boldsymbol{k}_1} P_1(\boldsymbol{k}_1, \boldsymbol{k}|s)$ according to equation (14) is the solution of the equation

$$\left[s + \frac{e\mathcal{E}}{\hbar}\nabla_{\boldsymbol{k}}\right]P_1(\boldsymbol{k}, s) - \sum_{\boldsymbol{k}'} P_1(\boldsymbol{k}', s)W(\boldsymbol{k}', \boldsymbol{k}|s)$$
$$= \frac{\mathrm{i}}{s}v_z(\boldsymbol{k})f(\boldsymbol{k}, s) + \frac{1}{s}\sum_{\boldsymbol{k}'} f(\boldsymbol{k}', s)W_1(\boldsymbol{k}', \boldsymbol{k}|s), \tag{24}$$

from which the sum rule

$$s^2\sum_{\boldsymbol{k}} P_1(\boldsymbol{k}, s) = \mathrm{i}\sum_{\boldsymbol{k}} v_{eff}(\boldsymbol{k}, s)f(\boldsymbol{k}, s) \tag{25}$$

is obtained. To construct a solution which accounts for this restriction and which elucidates the singular behaviour of $P_1(\boldsymbol{k}, s)$ with respect to s we make the ansatz

$$P_1(\boldsymbol{k}, s) = \frac{\mathrm{i}}{s}\varphi(\boldsymbol{k}, s) + \frac{1}{s^2}B(s)f(\boldsymbol{k}, s), \tag{26}$$

with the constraint

$$\sum_{\boldsymbol{k}} \varphi(\boldsymbol{k}, s) = 0. \tag{27}$$

The function $B(s)$ is completely determined by the sum rule (25). We obtain

$$B(s) = \mathrm{i}\sum_{\boldsymbol{k}} v_{eff}(\boldsymbol{k}, s)f(\boldsymbol{k}, s) = \mathrm{i}v_z(s). \tag{28}$$

$\varphi(\boldsymbol{k}, s)$ is the remaining unknown function, which we have to calculate. Inserting the ansatz of equation (26) together with (28) into (23), we obtain from equation (4) our final result for the diffusion coefficient

$$D_{zz}(s) = \sum_{\boldsymbol{k}} v_{eff}(\boldsymbol{k}, s)\varphi(\boldsymbol{k}, s) - \tfrac{1}{2}\sum_{\boldsymbol{k},\boldsymbol{k}'} f(\boldsymbol{k}, s)W_2(\boldsymbol{k}, \boldsymbol{k}'|s), \tag{29}$$

in which the s dependence is regular and the limit $s \to 0$ can easily be carried out. This is the most general result that describes carrier diffusion in semiconductors on which an electric field of arbitrary strength is applied. The second contribution on the right-hand side of equation (29) vanishes when the dipole operator commutes with the Hamiltonian. In this case, the expression for the diffusion coefficient corresponds to the one for the drift velocity. However, the function

$\varphi(\boldsymbol{k}, s=0)$ does not have the meaning of a carrier distribution function as does the function $f(\boldsymbol{k}, s=0)$ in equation (21). The kinetic equation for $\varphi(\boldsymbol{k}, s)$ is obtained from equation (24)

$$\frac{e\mathcal{E}}{\hbar}\frac{\partial}{\partial k_z}\varphi(\boldsymbol{k}, s) = \sum_{\boldsymbol{k}'}\varphi(\boldsymbol{k}', s)W(\boldsymbol{k}', \boldsymbol{k}|s) + v_z(\boldsymbol{k})f(\boldsymbol{k}, s) - \sum_{\boldsymbol{k}'}v_{eff}(\boldsymbol{k}', s)f(\boldsymbol{k}', s) - \mathrm{i}\sum_{\boldsymbol{k}'}f(\boldsymbol{k}', s)W_1(\boldsymbol{k}', \boldsymbol{k}|s). \tag{30}$$

This equation remains inhomogeneous even in the limit $s \to 0$, whereas equation (20) for the distribution function becomes homogeneous. In contrast to the average drift velocity, it is not sufficient to obtain a solution for the carrier distribution function in order to calculate the diffusion coefficient. Rather, it is necessary to determine a completely new quantity, which cannot be identified with a distribution function and which is the solution of equation (30). In the limit of weak electric fields, however, the Einstein relation is recovered, which relates the diffusion coefficient to the drift velocity (mobility) in a simple manner. To derive the Einstein relation for low electric fields, let us treat the standard situation when the Hamiltonian commutes with the dipole operator. Taking into account only the lowest-order contributions in the electric field, we obtain from equations (21) and (29)

$$v_z^{(1)} = \sum_{\boldsymbol{k}} v_z(\boldsymbol{k})f^{(1)}(\boldsymbol{k}), \tag{31}$$

$$D_{zz}^{(0)} = \sum_{\boldsymbol{k}} v_z(\boldsymbol{k})\varphi^{(0)}(\boldsymbol{k}), \tag{32}$$

where the superscript (1) or (0) refers to the order in the electric field. As has been rigorously shown in [9], there is a close relationship between $f^{(1)}(\boldsymbol{k})$ and $\varphi^{(0)}(\boldsymbol{k})$ expressed by

$$f^{(1)}(\boldsymbol{k}) = \frac{e\mathcal{E}}{k_B T}\varphi^{(0)}(\boldsymbol{k}). \tag{33}$$

Inserting this equation into (31), we obtain for the mobility $\mu = v_z^{(1)}/\mathcal{E}$ the Einstein relation

$$\mu = \frac{eD_{zz}}{k_B T}. \tag{34}$$

At the end of this section, we want to point out that the derivative W_1 of the scattering probability enters equation (30) even when the dipole operator commutes with the Hamiltonian. It is just this term that plays an essential role in the description of quantum diffusion in the WS regime. This observation leads us to the main physical conclusion of our paper. The derivative $W_1(\boldsymbol{k}', \boldsymbol{k}|s) = \partial W(\boldsymbol{k}', \boldsymbol{k}, \boldsymbol{\kappa}|s)/\partial\kappa_z|_{\boldsymbol{\kappa}=0}$, which enters the collision integral, refers to spatially inhomogeneous properties of the system. This inhomogeneity is an inherent feature of diffusion processes which we do not encounter in the treatment of the drift velocity. The description of diffusion phenomena requires the development of a separate theory, in which, contrary to the drift velocity, the carrier distribution function does not play a central role. Only in some limiting cases, when the Einstein relation is satisfied, do both quantities have the same origin.

3. Quantum diffusion in the Wannier–Stark regime

In this section the general expression for the diffusion coefficient derived in the previous section is written in an equivalent form, from which the main contributions for a given electric-field strength are easily identified. We start from our general result for the diffusion coefficient in equation (29) and apply integration by parts. The resulting quantity $\partial\varphi/\partial k_z$ is replaced by the right-hand side of equation (30), in which $\varphi(\boldsymbol{k})$ is substituted by the function

$$\Phi(\boldsymbol{k}) = \varphi(\boldsymbol{k}) - \frac{1}{e\mathcal{E}}\epsilon(\boldsymbol{k})f(\boldsymbol{k}), \qquad \epsilon(\boldsymbol{k}) = \varepsilon(\boldsymbol{k}) - \sum_{k_z}\varepsilon(\boldsymbol{k}). \tag{35}$$

Taking the limit $s \to 0$, our general result in equation (29) for the diffusion coefficient is expressed by

$$D_{zz} = \frac{1}{2}\frac{1}{(e\mathcal{E})^2}\sum_{\boldsymbol{k},\boldsymbol{k}'}[\epsilon(k_z)-\epsilon(k_z')]^2 f(\boldsymbol{k}')W(\boldsymbol{k}',\boldsymbol{k}) + \frac{\mathrm{i}}{e\mathcal{E}}\sum_{\boldsymbol{k},\boldsymbol{k}'}[\epsilon(k_z)-\epsilon(k_z')]f(\boldsymbol{k}')W_1(\boldsymbol{k}',\boldsymbol{k})$$
$$+\frac{1}{e\mathcal{E}}\sum_{\boldsymbol{k},\boldsymbol{k}'}[\epsilon(k_z)-\epsilon(k_z')]\Phi(\boldsymbol{k}')W(\boldsymbol{k}',\boldsymbol{k})$$
$$-\mathrm{i}\sum_{\boldsymbol{k},\boldsymbol{k}'}\varphi(\boldsymbol{k}')W_1(\boldsymbol{k}',\boldsymbol{k}) - \tfrac{1}{2}\sum_{\boldsymbol{k},\boldsymbol{k}'}f(\boldsymbol{k}')W_2(\boldsymbol{k}',\boldsymbol{k}), \tag{36}$$

which still provides an exact description of the field-dependent diffusion coefficient. The last two terms on the right-hand side of this equation disappear in the standard situation, when the interaction part of the Hamiltonian does not depend on the kinetic energy of the carriers so that $\sum_{\boldsymbol{k}} W_{1,2}(\boldsymbol{k}',\boldsymbol{k}) = 0$. We want to point out that there are cases where just these contributions dominate, as, for example, in the theory of small polarons. The third term on the right-hand side of equation (36) is irrelevant for high electric fields, when $\Omega\tau \gtrsim 1$ is satisfied (Ω denotes the Bloch frequency and τ an effective scattering time). This can be seen by an analysis of the kinetic equation for $\Phi(\boldsymbol{k})$, which is readily obtained from equations (30) and (35). The result $\Phi \sim \mathcal{E}^{-3}$ leads to a contribution to the diffusion coefficient, which is proportional to $\mathcal{E}^{-4}$. For intermediate field strengths (quasi-classical limit), when the Stark ladder is not fully resolved $\Omega\tau \sim 1$ and the band width Δ is sufficiently large so that $\Delta/e\mathcal{E}d > 1$ is still satisfied, the most important contribution to the diffusion coefficient is given by the first term on the right-hand side of equation (36) with a field-independent scattering probability. This conclusion is justified by the physical picture presented in appendix A. When the electric field increases further so that the quantum regime is reached where WS states are formed, all terms on the right-hand side of equation (36) become relevant except the contribution proportional to Φ. This regime of quantum diffusion is most suitably described within the Houston representation, in which these contributions can be combined to a compact expression for the diffusion coefficient. The details of the calculation are presented in appendix B. The result

$$D_{zz} = \tfrac{1}{2}\sum_{\boldsymbol{k}_\perp,\boldsymbol{k}_\perp'}\sum_{l=-\infty}^{\infty}(ld)^2 f(\boldsymbol{k}_\perp')W_{0l}^{0l}(\boldsymbol{k}_\perp',\boldsymbol{k}_\perp) \tag{37}$$

allows a clear physical interpretation within the hopping picture, in which quantum diffusion is due to inelastic scattering. The diffusion coefficient is composed of the square of the hopping length $(ld)^2$ (with l being the layer index), the lateral carrier distribution function $f(\boldsymbol{k}_\perp)$ and the scattering probabilities in the site representation. For both classically high electric fields and the WS regime the diffusion coefficient is completely determined by the scattering probability and the carrier distribution function. The quantum-diffusion picture expressed by equation (37) is reminiscent of the expression for the drift velocity valid for quantizing electric fields [10]. By switching to the Houston representation, we obtain from equations (21) and (22)

$$v_z = \sum_{\boldsymbol{k}_\perp,\boldsymbol{k}_\perp'}\sum_{l=-\infty}^{\infty}(ld)f(\boldsymbol{k}_\perp')W_{0l}^{0l}(\boldsymbol{k}_\perp',\boldsymbol{k}_\perp), \tag{38}$$

which is interpreted within the hopping transport picture as an average shift of the carriers per time interval (note that the scattering probability W has the dimension s^{-1}). The expressions in equations (37) and (38) are exact in the WS regime, when the WS ladder is fully developed ($\Omega\tau > 1$) so that quantum effects become most prominent. Most interesting are electron–phonon resonances, which have been identified in the drift velocity of narrow-band semiconductors [11] and SLs [12], and which, according to equations (37) and (38), also

appear in the diffusion coefficient with the same nonanalytic lineshape and at the same field strengths.

In the ultra-quantum limit ($\Omega\tau \gg 1$), there is only hopping between nearest neighbour layers (i.e. the layer index l is restricted to -1, 0 and 1). In this regime, the lateral distribution function has the form $f(\boldsymbol{k}_\perp) \sim \exp[-\varepsilon(\boldsymbol{k}_\perp/k_BT)]$ [11]. By taking into account the principle of detailed balance for transitions between nearest-neighbour layers in an external field

$$W_{01}^{01}(\boldsymbol{k}'_\perp, \boldsymbol{k}_\perp)/\, W_{0-1}^{0-1}(\boldsymbol{k}_\perp, \boldsymbol{k}'_\perp) = \exp\left(\frac{e\mathcal{E}d + \varepsilon(\boldsymbol{k}'_\perp) - \varepsilon(\boldsymbol{k}_\perp)}{k_BT}\right), \tag{39}$$

the expressions for the diffusion coefficient and the drift velocity as obtained from equations (37) and (38) can be related to each other. The result

$$D_{zz} = \frac{v_z d}{2} \coth \frac{e\mathcal{E}d}{2k_BT}, \tag{40}$$

which is valid in the ultra-quantum limit ($\Omega\tau \gg 1$), preserves the symmetry of the diffusion coefficient with respect to the electric field. Whereas the drift velocity is antisymmetric $v_z(-\mathcal{E}) = -v_z(\mathcal{E})$, we obtain from equation (40) for the diffusion coefficient the symmetry property $D_{zz}(-\mathcal{E}) = D_{zz}(\mathcal{E})$. If in addition to $\Omega\tau \gg 1$, the inequality $2k_BT < e\mathcal{E}d$ is satisfied, both the drift velocity and the diffusion coefficient decrease with increasing electric field in the same manner. The detailed form of the negative differential conductivity, which is expected to occur in the hopping transport regime, depends on the relevant scattering mechanism [13]. When scattering on polar-optical phonons dominates, the drift velocity exhibits a $1/\mathcal{E}^2$ dependence in the ultra-quantum limit. Other scattering mechanisms give rise to other characteristic field asymptotics. For the mobility $\mu = v_z/\mathcal{E}$, we obtain from equation (40)

$$\mu = \frac{eD_{zz}}{k_BT} \frac{\tanh(e\mathcal{E}d/2k_BT)}{e\mathcal{E}d/2k_BT}, \tag{41}$$

which reproduces the Einstein relation in the limit of sufficiently high temperatures ($e\mathcal{E}d < 2k_BT$). Under the condition $2k_BT < e\mathcal{E}d$, we have $\mu = 2D_{zz}/(\mathcal{E}d)$, which corresponds to $D_{zz} = v_z d/2$.

4. Summary

A general microscopic theory has been developed for the carrier diffusion in semiconductors and semiconductor SLs, at which a constant electric field of arbitrary strength is applied. The approach has been restricted to the treatment of one occupied band. Unlike the drift velocity or the current density, the diffusion coefficient results from a function that does not have the meaning of a distribution function. It is necessary to derive a specific equation for this quantity, which has been accomplished in section 2. In both the drift velocity and the diffusion coefficient there is an additional scattering-induced contribution when the Hamiltonian does not commute with the dipole operator. This happens, for example, in the theory of small polarons. Our final result as expressed in equations (29) and (37) allows a detailed study of field-mediated diffusion phenomena in realistic three-dimensional semiconductor (or SL) models. First results have already been obtained for a one-dimensional SL model treated in the quasi-elastic scattering regime [14]. Further applications may start from the kinetic equation (30) (which plays the same role for the diffusion coefficient as the Boltzmann equation for the drift velocity) and the representation (29), which has a similar form to equation (21) for the drift velocity. Such an approach is applicable for systems with delocalized electronic states. The treatment of the WS regime, however, would start from the Houston representation (37), which describes quantum

diffusion in the hopping picture. A characteristic effect of quantum diffusion is the appearance of electron–phonon resonances, which were predicted many years ago to occur in the current density [15]. From the general results for the drift velocity in equation (38) and the diffusion coefficient in equation (37), we conclude that the energy positions and the nonanalytic form of the electron–phonon resonances agree in both quantities. In the ultra-quantum limit ($\Omega\tau \gg 1$), the diffusion coefficient exhibits the same electric-field dependence as the drift velocity.

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Appendix A. Diffusion at classically high electric fields

On the basis of physical arguments, the diffusion coefficient for classically high electric fields is considered. Let us treat the hopping-like motion of carriers induced by scattering as illustrated in figure A.1. The particle remains at a given position z_i until it hops due to scattering to another one. After n steps, the particle reaches the position

$$R_n = \sum_{i=1}^{n} \Delta z_i. \tag{A.1}$$

The diffusion coefficient is related to R_n^2. The time evolution of this quantity has the form

$$R(t)^2 = \sum_{n=0}^{\infty} R_n^2 \int_0^t \mathrm{d}t' \,[\delta(t' - \tau_1 - \cdots - \tau_n) - \delta(t' - \tau_1 - \cdots - \tau_n - \tau_{n+1})]. \tag{A.2}$$

To calculate $D_{zz}(s)$, we need the Laplace transform of $R(t)^2$, which is expressed by

$$Z_2(s) = \frac{1}{s}\int_0^\infty \mathrm{d}t\, \mathrm{e}^{-st} \frac{\mathrm{d}R(t)^2}{\mathrm{d}t} = \frac{1}{s}\sum_{n=0}^{\infty} R_n^2 \mathrm{e}^{-s(\tau_1+\cdots+\tau_n)}(1 - \mathrm{e}^{-s\tau_{n+1}}), \tag{A.3}$$

where an integration by parts has been applied. To facilitate the consideration, the required time average is performed over statistically independent time steps. We obtain

$$\langle Z_2(s)\rangle = \frac{1}{s}\sum_{n=0}^{\infty}\left\{n\lambda^{n-1}(1-\lambda)\langle \mathrm{e}^{-s\tau}(\Delta z)^2\rangle + n(n-1)\lambda^{n-2}(1-\lambda)\langle \Delta z \mathrm{e}^{-s\tau}\rangle^2\right\}, \tag{A.4}$$

with $\lambda = \langle \exp(-s\tau)\rangle$. In the limit $t \to \infty$ (or $s \to 0$), we expand all quantities with respect to s and retain only the most divergent contributions to $\langle Z_2(s)\rangle$. The result

$$\langle Z_2(s)\rangle \approx \frac{1}{s^2}\frac{\langle(\Delta z)^2\rangle}{\langle\tau\rangle} + \frac{2}{s^3}\left(\frac{\langle\Delta z\rangle}{\langle\tau\rangle}\right)^2 \tag{A.5}$$

is expressed by the mean values $\langle\Delta z\rangle$ and $\langle(\Delta z)^2\rangle$, which are estimated taking into account the field-induced periodic carrier motion [11]. For a one-dimensional system, the velocity of this periodic motion is given by

$$v[k_z(t)] = \frac{1}{\hbar}\frac{\mathrm{d}\varepsilon(k_z + e\mathcal{E}t/\hbar)}{\mathrm{d}k_z} = \frac{1}{e\mathcal{E}}\frac{\mathrm{d}\varepsilon}{\mathrm{d}t}, \tag{A.6}$$

where the dispersion relation is periodic $\varepsilon(k_z + 2\pi/d) = \varepsilon(k_z)$. Due to scattering, the quasi momentum of the carrier changes from k_z to k_z' at a given moment T. Afterwards the carrier oscillates again in k space and takes over the original quasi momentum k_z at a later time $T + \Delta t$. During the time interval Δt, the carrier is displaced by

$$\Delta z = \int_T^{T+\Delta t} \mathrm{d}t\, v[k_z(t)] = \frac{\varepsilon(k_z) - \varepsilon(k_z')}{e\mathcal{E}}, \tag{A.7}$$

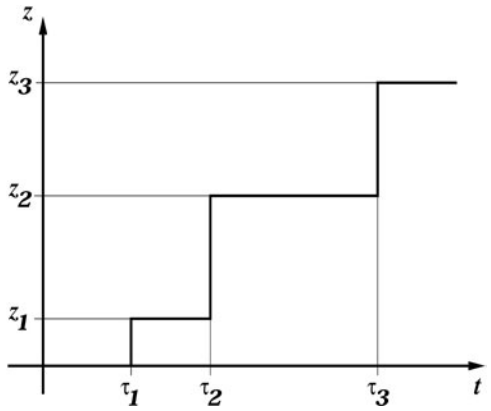

Figure A.1. Schematic diagram of the trajectory of a classical particle in the space–time coordinate system.

which is averaged by using the scattering probability. For a three-dimensional system, we introduce the lateral distribution function and obtain for the scattering-induced carrier shift

$$\langle \Delta z \rangle = \sum_{\boldsymbol{k},\boldsymbol{k}'} \frac{\epsilon(k_z) - \epsilon(k_z')}{e\mathcal{E}} f(\boldsymbol{k}_\perp') W(\boldsymbol{k}', \boldsymbol{k}) \langle \tau \rangle. \tag{A.8}$$

Accordingly, we obtain

$$\langle (\Delta z)^2 \rangle = \sum_{\boldsymbol{k},\boldsymbol{k}'} \left(\frac{\epsilon(k_z) - \epsilon(k_z')}{e\mathcal{E}} \right)^2 f(\boldsymbol{k}_\perp') W(\boldsymbol{k}', \boldsymbol{k}) \langle \tau \rangle, \tag{A.9}$$

which is inserted into equation (A.5). The final result for the diffusion coefficient is obtained from equation (4)

$$D_{zz} = \frac{1}{2} \frac{1}{(e\mathcal{E})^2} \sum_{\boldsymbol{k},\boldsymbol{k}'} (\epsilon(k_z) - \epsilon(k_z'))^2 f(\boldsymbol{k}_\perp') W(\boldsymbol{k}', \boldsymbol{k}), \tag{A.10}$$

where $v_z = \langle z \rangle / \langle \tau \rangle$ has been used. The expression for the diffusion coefficient in equation (A.10), derived for quasi-classical particles subject to strong electric fields, completely agrees with the first term on the right-hand side of equation (36). This result can be compared with the expression for the drift velocity $v_z = \langle z \rangle / \langle \tau \rangle$ obtained from equation (A.8). In the considered regime of classically high electric fields, the drift velocity decreases with increasing field according to $v_z \sim 1/\mathcal{E}$, whereas the diffusion coefficient exhibits an inverse quadratic dependence $D_{zz} \sim 1/\mathcal{E}^2$. For quasi-elastic scattering, we again recover the Einstein relation, when the principle of detailed balance is taken into account.

Appendix B. The scattering probability in the WS representation

In this appendix, we will exploit the Houston representation for the scattering probability [11]

$$\begin{aligned} W(\boldsymbol{k}', \boldsymbol{k}, \boldsymbol{\kappa}) = \sum_{m_i} W_{m_2 m_4}^{m_1 m_3}(\boldsymbol{k}_\perp', \boldsymbol{k}_\perp) \exp\Big\{ & \mathrm{i}(m_3 - m_4) k_z d - \mathrm{i}(m_1 - m_2) k_z' d \\ & + \frac{\mathrm{i}}{2}(m_1 + m_2 - m_3 - m_4)\kappa_z d - \mathrm{i}\chi\left(\boldsymbol{k}' + \frac{\boldsymbol{\kappa}}{2}\right) + \mathrm{i}\chi\left(\boldsymbol{k}' - \frac{\boldsymbol{\kappa}}{2}\right) \\ & + \mathrm{i}\chi\left(\boldsymbol{k} + \frac{\boldsymbol{\kappa}}{2}\right) - \mathrm{i}\chi\left(\boldsymbol{k} - \frac{\boldsymbol{\kappa}}{2}\right) \Big\}, \end{aligned} \tag{B.1}$$

with $\chi(\boldsymbol{k})$ being determined by

$$e\mathcal{E}\frac{\partial\chi(\boldsymbol{k})}{\partial k_z} = \epsilon(\boldsymbol{k}), \tag{B.2}$$

to derive the final expression (37) for the quantum diffusion. We start from equation (29) for the diffusion coefficient, perform an integration by parts, and replace the quantity $\partial\varphi/\partial k_z$ by the right-hand side of the kinetic equation (30). Taking into account the relation of detailed balance $\sum_{\boldsymbol{k}} W(\boldsymbol{k}', \boldsymbol{k}) = 0$ and calculating the quantities $W_1(\boldsymbol{k}', \boldsymbol{k})$ and $W_2(\boldsymbol{k}', \boldsymbol{k})$ from equation (B.1), we obtain

$$\begin{aligned} D_{zz} = \frac{d}{2}\sum_{\boldsymbol{k},\boldsymbol{k}'}\Phi(\boldsymbol{k}')\sum_{m_i}(m_1+m_2-m_3-m_4)W^{m_1m_3}_{m_2m_4}(\boldsymbol{k}'_\perp, \boldsymbol{k}_\perp)\mathrm{e}^{-\mathrm{i}(m_1-m_2)k'_zd+\mathrm{i}(m_3-m_4)k_zd} \\ + \frac{d^2}{2}\sum_{\boldsymbol{k},\boldsymbol{k}'}f(\boldsymbol{k}')\sum_{m_i}\frac{1}{4}(m_1+m_2-m_3-m_4)^2W^{m_1m_3}_{m_2m_4}(\boldsymbol{k}'_\perp, \boldsymbol{k}_\perp) \\ \times\,\mathrm{e}^{-\mathrm{i}(m_1-m_2)k'_zd+\mathrm{i}(m_3-m_4)k_zd}. \end{aligned} \tag{B.3}$$

At high electric fields ($\Omega\tau > 1$), the first contribution, which is proportional to Φ, can be neglected. In the second contribution on the right-hand side of equation (B.3), $f(\boldsymbol{k})$ is replaced by the lateral distribution function $f(\boldsymbol{k}_\perp)$. After carrying out the k_z and k'_z integrals, we arrive at the final result in equation (37), valid in the Stark ladder regime ($\Omega\tau > 1$).

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