# Nonlinear transport of polarons

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A method is presented to calculate the nonlinear mobility of a Fröhlich polaron in high electric fields for arbitrary strength of the electron-phonon coupling. It is based on an eikonal expansion of the exact Feynman influence functional for the nonequilibrium Wigner distribution function. The method becomes exact both in linear response and for large transport velocities. The nonlinear mobility is obtained from a momentum balance equation containing a systematic frictional force and the field-dependent fluctuations due to the eliminated phonon degrees of freedom. For weak coupling we recover the standard results of Stratton and Conwell. For strong coupling, however, the current-voltage characteristics are quite different from those obtained by Thornber and Feynman. Our results are in good agreement with measurements of high-field transport in InSb and AgCl.

## I. INTRODUCTION

The nonlinear transport of electrons in polar semiconductors or ionic crystals is a subject of long-standing interest.[1] The basic model to describe the dominant phonon scattering in an otherwise perfect lattice is due to Fröhlich.[2] There an electron couples to the linear displacement induced by longitudinal optical phonons. In spite of the single-particle nature of this model, even the equilibrium problem cannot be solved exactly unless the coupling is treated in second-order perturbation theory. Therefore a major breakthrough was the Feynman path-integral formulation.[3] It allowed one to eliminate the phonon degrees of freedom exactly for arbitrary coupling, leading to an effective single particle with a retarded self-interaction. For equilibrium properties like the ground-state energy or the effective mass, this method proved to be extremely powerful. Indeed, while the exact path integral is not soluble, a simple variational ansatz in which the electron couples to an effective single oscillator turned out to give excellent results for arbitrary values of the coupling strength. In a nonequilibrium steady state, however, which occurs in a transport situation with a finite electric field, there is no variational principle allowing one to determine the parameters of the effective oscillator model. As a result, Feynman *et al.*[4] and Thornber and Feynman[5] in their treatment of linear and nonlinear polaron transport were forced to assume that these parameters retain their equilibrium values even in the presence of a finite external field, at least in the reference frame moving with the electron on average. Clearly, such an assumption is difficult to justify. The aim of our present work is therefore to develop a method which does not rely on applying equilibrium properties in a transport situation. It is essentially based on the eikonal method in scattering theory where the exact propagators are evaluated at a path moving with a definite drift velocity. Specifically a Gaussian expansion around a given average motion of the center-of-mass coordinate allows one to evaluate the exact collision term of the quantum-mechanical Wigner distribution in a steady state. As expected, this method becomes exact for large transport velocities. In turns out, however, that it is also correct in the linear response regime at least for small coupling. Thus we expect that our approximation is reliable for arbitrary fields. This is supported by the fact that experimental data on the nonlinear mobility in InSb and AgCl are in good agreement with our results.

The paper is organized as follows: In Sec. II we use the Feynman-Vernon influence functional[6] to derive a formally exact equation of motion for the time-dependent Wigner distribution of an electron coupled to optical phonons within the Fröhlich model. The associated momentum balance in a steady state requires the calculation of a complicated double path integral. Using an eikonal approximation, the problem is reduced to the evaluation of conventional integrals. It is shown explicitly that the asymptotic steady state is independent of the precise initial condition. In Sec. III the momentum balance is first evaluated analytically for weak coupling. There it turns out that our results coincide with those of Stratton[7] and Conwell,[8] which are generalized to the case of a finite Brillouin zone. Measurements of the nonlinear mobility in the weakly polar semiconductor InSb covering more than two orders of magnitude are shown to be in very good agreement with our results. In the strong-coupling case, the implicit relation between current and field is determined numerically. The results deviate strongly from those obtained by Thornber and Feynman. In particular, the current-voltage characteristics are highly nonlinear at the threshold for single-phonon emission, which is determined by the bare mass even for large coupling. Qualitative agreement is obtained with measurements on the high-field mobility in AgCl. Section IV contains a brief discussion of our results and their relation to previous and alternative treatments. Finally, a

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discussion of the well-known $\frac{3}{2}k_BT$ problem in the linear mobility is given in the Appendix.

## II. INFLUENCE FUNCTIONAL THEORY

### A. General formalism

We start from the standard Fröhlich model in which a single electron with band mass $M$ is coupled to longitudinal optical phonons of a harmonic lattice. Taking the phonon frequency $\omega_0$ to be independent of momentum $\vec{k}$, the unperturbed Hamiltonian is

$$H_0=\frac{\vec{p}^2}{2M}+\hbar\omega_0\sum_k a_k^\dagger a_k \ , \tag{1}$$

with the usual phonon creation and annihilation operators $a_k^\dagger$ and $a_k$. The coupling to the linear dimensionless displacement,

$$x_k=a_k+a_{-k}^\dagger \ , \tag{2}$$

associated with the ionic polarizability, leads to an interaction of the form

$$H_I=V^{-1/2}\sum_k \lambda_k \exp(i\vec{k}\cdot\vec{q})x_k \ . \tag{3}$$

Here $V\to\infty$ is the volume, $\vec{q}$ the electron coordinate, and

$$\lambda_k=\frac{\lambda}{|\vec{k}|} \tag{4}$$

the coupling constant. The parameter $\lambda$ is related to the standard Fröhlich coupling constant[2]

$$\alpha=\frac{e^2}{\hbar}\left[\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right]\left[\frac{M}{2\hbar\omega_0}\right]^{1/2} \tag{5}$$

by

$$\lambda=-(4\pi\alpha)^{1/2}\hbar\omega_0\left[\frac{\hbar}{2M\omega_0}\right]^{1/4} \ . \tag{6}$$

In the presence of a uniform and static electric field $\vec{E}=\vec{F}/e$ $(e>0)$, the total Hamiltonian is

$$H=H_0+H_I-\vec{F}\cdot\vec{q} \ . \tag{7}$$

Since we are interested in nonequilibrium properties and moreover want to eliminate the phonons, we introduce the reduced time-dependent matrix $\rho(t)=\mathrm{Tr}_B\rho_{\mathrm{tot}}(t)$ as a trace over the bath degrees of freedom. As has been shown by Feynman and Vernon,[6] its position space matrix elements can be calculated from their initial value $\langle\vec{q}_0|\rho(0)|\vec{q}_0'\rangle$ via

$$\langle\vec{q}|\rho(t)|\vec{q}'\rangle=\int d^3q_0 d^3q_0'\langle\vec{q}_0|\rho(0)|\vec{q}_0'\rangle J(\vec{q},\vec{q}',t;\vec{q}_0,\vec{q}_0') \ , \tag{8}$$

provided that the total density matrix $\rho_{\mathrm{tot}}(0)=\rho(0)\rho_B(0)$ factorizes initially. The associated propagator

$$J=\int_{\vec{q}_0}^{\vec{q}}D^3q\int_{\vec{q}_0'}^{\vec{q}'}D^3q'^*\exp\left\{\frac{i}{\hbar}(S[\vec{q}]-S[\vec{q}'])\right\}\cdot F[\vec{q},\vec{q}'] \tag{9}$$

is written as a double path integral with the classical action

$$S[\vec{q}]=\int_0^t dt_1\left[\frac{M}{2}\dot{\vec{q}}^2+\vec{F}\cdot\vec{q}\right] \tag{10}$$

of the uncoupled electron. Here $D^3q$ is the usual Feynman differential for discretized paths,

$$D^3q=\lim_{N\to\infty}\left[\frac{M}{2\pi\hbar i\Delta t}\right]^{3N/2}\prod_{j=1}^{N-1}d^3q_j \ , \quad \Delta t=t/N \ . \tag{11}$$

Moreover, the elimination of the phonons leads to a nontrivial influence functional $F[\vec{q},\vec{q}']$, which couples the paths $\vec{q}(t_1)$ and $\vec{q}'(t_1)$.[6] Defining an expectation value for the phonon degrees of freedom with their initial equilibrium density matrix as $\langle\ldots\rangle=\mathrm{Tr}_B[\ldots\rho_B(0)]$, the influence functional can quite generally be written as

$$F[\vec{q},\vec{q}']=\langle U_{\vec{q}'}^\dagger U_{\vec{q}}\rangle \ . \tag{12}$$

Here $U_{\vec{q}}$ is the unitary time evolution operator for the Hamiltonian $H_B+H_I[\vec{q}]$ in which a given electron path $\vec{q}(t_1)$ acts as a $c$-number source for the phonon system $H_B$. Introducing the influence phase $\phi$ by $F[\vec{q},\vec{q}']=\exp\{i\phi[\vec{q},\vec{q}']\}$ and the equilibrium correlation function of the phonon-induced displacements,

$$S(t)=\langle x_k(t)x_{-k}\rangle=\coth\frac{\beta\hbar\omega_0}{2}\cos\omega_0 t-i\sin\omega_0 t \ , \tag{13}$$

the influence phase arising from the driven oscillator system is known exactly as[6]

$$i\phi=-\frac{1}{\hbar^2}\int_0^t dt_1\int_0^{t_1}dt_2\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2[e^{i\vec{k}\cdot\vec{q}(t_1)}-e^{i\vec{k}\cdot\vec{q}'(t_1)}][e^{-i\vec{k}\cdot\vec{q}(t_2)}S(t_1-t_2)-e^{-i\vec{k}\cdot\vec{q}'(t_2)}S^*(t_1-t_2)] \ . \tag{14}$$

It is convenient to introduce the center-of-mass and relative coordinates

$$\vec{x}=(\vec{q}+\vec{q}')/2\ ,\quad \vec{y}=\vec{q}-\vec{q}'\ , \tag{15}$$

and define a function

$$\langle \vec{x}+\vec{y}/2|\rho(t)|\vec{x}-\vec{y}/2\rangle=\rho(\vec{x},\vec{y},t)\ . \tag{16}$$

Its Fourier transform with respect to the off-diagonal elements $\vec{y}$,

$$f(\vec{x},\vec{p},t)=\int \frac{d^3y}{(2\pi\hbar)^3}\rho(\vec{x},\vec{y},t)e^{-i\vec{p}\cdot\vec{y}/\hbar}\ , \tag{17}$$

is then precisely the Wigner distribution function which generalizes the classical phase space distribution to a quantum system. Similarly to the classical case, this function obeys an equation of motion of the form[9,10]

$$\left[\partial_t+\frac{\vec{p}}{M}\partial_{\vec{x}}+\vec{F}\partial_{\vec{p}}\right]f(\vec{x},\vec{p},t)=[\partial_t f(\vec{x},\vec{p},t)]_{\rm coll}\ . \tag{18}$$

This may be derived by considering the double propagator $J$ for an infinitesimal time interval between $t$ and $t+dt$, similar to the derivation of the Schrödinger equation from the standard path-integral representation of the single propagator. Formally, (18) has the form of a one-particle Boltzmann equation; however, the collision term

$$[\partial_t f]_{\rm coll}=\int \frac{d^3y}{(2\pi\hbar)^3}e^{-i\vec{p}\cdot\vec{y}/\hbar}\int d^3x_0 d^3y_0\rho(\vec{x}_0,\vec{y}_0)$$
$$\times\int_{\vec{x}_0}^{\vec{x}}D^3x\int_{\vec{y}_0}^{\vec{y}}D^3y\exp\left\{\frac{i}{\hbar}\int_0^t dt_1(M\dot{\vec{x}}\cdot\dot{\vec{y}}+\vec{F}\cdot\vec{y})+i\phi[\vec{x},\vec{y}]\right\}\partial_t i\phi[\vec{x},\vec{y}] \tag{19}$$

is extremely complicated, depending explicitly on the initial distribution and all times between 0 and $t$. It can be shown that the standard Boltzmann collision term is obtained from (19) by evaluating the factor $\partial_t i\phi$ at ballistic paths $\vec{x}(t)=\vec{x}_0+t\vec{p}/M$.[10]

In principle, Eq. (18) provides a complete description of the nonequilibrium dynamics of a single electron where the phonons have been eliminated exactly for arbitrary coupling and which still contains the full quantum-mechanical behavior. For the case of high-field dc transport, it is sufficient to consider time-independent steady-state solutions. Multiplying Eq. (18) with $\vec{p}$ and integrating, we obtain an exact momentum balance condition

$$-\vec{F}=\int d^3x\,d^3p\,\vec{p}\lim_{t\to\infty}[\partial_t f(\vec{x},\vec{p},t)]_{\rm coll}\ , \tag{20}$$

which is the basis for determining the nonlinear current-voltage characteristic. Clearly, in order to obtain explicit results, we need to evaluate the double path integral in (19). To this end it is convenient to split the influence phase $\phi$ into real and imaginary parts via

$$i\phi=i\phi_1-\phi_2\ . \tag{21}$$

Then, expressing everything in terms of $\vec{x}$ and $\vec{y}$, we obtain from (14)

$$\phi_1=\frac{4}{\hbar^2}\int_0^t dt_1\int_0^{t_1}dt_2\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\sin\vec{k}[\vec{x}(t_1)-\vec{x}(t_2)]\sin\frac{\vec{k}\cdot\vec{y}(t_1)}{2}\cos\frac{\vec{k}\cdot\vec{y}(t_2)}{2}\,{\rm Im}S(t_1-t_2) \tag{22}$$

and

$$\phi_2=\frac{4}{\hbar^2}\int_0^t dt_1\int_0^{t_1}dt_2\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\cos\vec{k}[\vec{x}(t_1)-\vec{x}(t_2)]\sin\frac{\vec{k}\cdot\vec{y}(t_1)}{2}\sin\frac{\vec{k}\cdot\vec{y}(t_2)}{2}\,{\rm Re}S(t_1-t_2)\ . \tag{23}$$

Because of the nonlinear trigonometric functions, the path integrals $D^3x$ and $D^3y$ cannot be calculated exactly. Therefore, in the following, we will discuss two simple approximations which allow one to determine the nonlinear current-voltage characteristic explicitly for arbitrary fields and coupling strength.

### B. Classical limit

As has been shown in the context of quantum dissipation,[11] the limit in which the electron may be treated classically is obtained by expanding the influence phase $\phi$ up to quadratic order in the off-diagonal elements $\vec{y}$ of the density matrix, which are formally of order $\hbar$. The resulting real and imaginary parts can then be written as

$$\phi_1^{\rm cl}=-\frac{1}{\hbar}\int_0^t dt_1\vec{y}(t_1)\vec{F}_B[\vec{x},t_1] \tag{24}$$

and

$$\phi_2^{\rm cl}=\frac{1}{\hbar^2}\int_0^t dt_1 y_\alpha(t_1)\int_0^{t_1} dt_2\langle \xi_\alpha[\vec{x},t_1]\xi_\beta[\vec{x},t_2]\rangle y_\beta(t_2) \tag{25}$$

($\alpha$ and $\beta$ are summed over). Here

$$\vec{F}_B[\vec{x},t_1]=-\frac{2}{\hbar}\int_0^{t_1} dt_2\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k}\sin\vec{k}[\vec{x}(t_1)-\vec{x}(t_2)]\,\mathrm{Im}S(t_1-t_2) \tag{26}$$

is the systematic frictional force which a classical electron "feels" as a result of its coupling to the lattice. The contribution $\phi_2$ in turn may be interpreted as the effect of an associated fluctuating force $\vec{\xi}[\vec{x},t]$. To see this, the factor $\exp(-\phi_2)$ is written as an average over a Gaussian stochastic process,[11,12]

$$e^{-\phi_2}=\left\langle\exp\left\{\frac{i}{\hbar}\int_0^t dt_1\vec{\xi}[\vec{x},t_1]\vec{y}(t_1)\right\}\right\rangle , \tag{27}$$

with a fluctuating force $\vec{\xi}$ which has zero average and variance

$$\langle\xi_\alpha[\vec{x},t_1]\xi_\beta[\vec{x},t_2]\rangle=\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2k_\alpha k_\beta\cos\vec{k}[\vec{x}(t_1)-\vec{x}(t_2)]\,\mathrm{Re}S(t_1-t_2) . \tag{28}$$

Note that both $\vec{F}_B$ and $\vec{\xi}$ are functionals of the diagonal path $\vec{x}(t_1)$. Since the off-diagonal element $\vec{y}$ now occurs only linearly [use (27)], the path integral $D^3y$ can easily be evaluated. Indeed, integrating the kinetic energy term $M\dot{\vec{x}}\cdot\dot{\vec{y}}$ by parts, its discretized version factorizes and leads to a product of $\delta$ functions which require that $\vec{x}(t)$ obey the classical generalized Langevin equation

$$M\ddot{\vec{x}}-\vec{F}+\vec{F}_B[\vec{x},t]=\vec{\xi}[\vec{x},t] . \tag{29}$$

Obviously, the frictional force is retarded and nonlinear in the velocity. Moreover, the Gaussian noise term does not have a white spectrum as is usually assumed (it is not even stationary in general) and in particular is also state dependent. It should also be pointed out that equations of the type (29) are sometimes called *quasiclassical*,[11] because the bath is still treated quantum mechanically and thus, e.g., the noise $\vec{\xi}[\vec{x},t]$ remains finite at $T=0$. Nevertheless, (29) does not contain quantum effects of the particle, like interference or tunneling, and thus is really an equation of motion for a classical particle in a quantum-mechanical environment.

In the context of high-field transport, equations of this type have previously been studied by several authors.[13,14] In particular, it has been realized that it is important to include the velocity fluctuations $\delta\dot{\vec{x}}$ in an expansion around the average path $\langle\vec{x}(t)\rangle=\vec{x}_0+\vec{v}t$. Indeed, if fluctuations are neglected, the associated frictional force

$$\begin{aligned}\vec{F}_B[\vec{x}=\vec{v}t,t\to\infty]&=\vec{F}_B(v)\\&=\frac{2}{\hbar}\int_0^\infty dt\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k}\sin\vec{k}\cdot\vec{v}\sin\omega_0 t ,\end{aligned} \tag{30}$$

and therefore the current-voltage relation obtained from the stationary averaged Langevin equation $\vec{F}=\vec{F}_B(v)$ will be completely *independent* of temperature. Moreover, for the Fröhlich model it turns out that

$$\vec{F}_B(v)=\frac{\lambda^2\omega_0}{2\pi\hbar v^2}\ln(v/\bar{v})\theta(v-\bar{v})\cdot\vec{e}_v \tag{31}$$

is only nonzero if the velocity is larger than $\bar{v}=\omega_0/k_{\max}$, where $k_{\max}>|\vec{k}|$ is the maximum $\vec{k}$ vector in the Brillouin zone. This is due to the fact that a *classical* electron with fixed velocity $\vec{v}$ interacts with the lattice only if the Landau condition $\vec{k}\cdot\vec{v}=\omega_0$ is obeyed, which is impossible to fulfill for $|\vec{k}|<k_{\max}$ if $v<\bar{v}$. Quantum mechanically, the threshold is at $\hbar\omega_0=Mv^2/2$; see below. In reality, of course, the velocity fluctuates, leading to a nonzero frictional force even for small $v$ and thus a finite mobility $\mu=v/F$ in the limit $v,F\to 0$.

The generalized Langevin equation (29) is only valid classically; however, it turns out that for large velocities it becomes exact quite generally. The reason for this may be seen from the general result (19) for the collision term. In fact, because of the factor $\exp[(i/\hbar)\vec{F}\int dt_1\vec{y}(t_1)]$, the double path integral for large $F$ may be calculated in a stationary-phase approximation around $\vec{y}=0$ since the main contribution comes from paths in which $\int dt_1\vec{y}$ is near zero. Thus quantum effects become irrelevant for large fields or transport velocities. It should be pointed out, however, that in this regime the Fröhlich model is strictly speaking, insufficient since the frictional force $\vec{F}_B(v)$ due to phonon scattering *decreases* with increasing velocity. As was already pointed out by Thornber and Feynman,[5] this prevents one from determining a unique inverse $v(F)$ from the calculated $F(v)$ and leads to a maximum field $F_c$ beyond which no steady state is possible. Moreover, the regime $v>v_c$, where $F(v)$ decreases with $v$, is unstable with respect to an unlimited increase in velocity. This problem can only be avoided if one includes umklapp processes or other additional sources of scattering like impact ionization at high fields.[15] It is interesting to see how the instability for $v>v_c$ shows up in the fluctuations around the steady state. From the explicit solution of a simple model, it maybe shown[16] that a generalized Langevin equation of the form (29) with a fric-

tional force which has negative slope for $v > v_c$ leads to diffusive fluctuations.

$$\langle (z(t)-vt)^2 \rangle = 2D_{\parallel}(v)t \tag{32}$$

and

$$\langle x^2(t) \rangle = \langle y^2(t) \rangle = 2D_{\perp}(v)t \tag{33}$$

around the average drift with anisotropic and state-dependent diffusion constants $D_{\parallel}(v) \neq D_{\perp}(v)$ (we have taken $\vec{F}$ and thus $\vec{v}$ in the $z$ direction). The instability of the motion for $v > v_c$ is revealed in the longitudinal diffusion constant $D_{\parallel}$, which diverges like $(v_c - v)^{-2}$ as $v$ approaches $v_c$ from below.[16] In the following we will develop a method to treat the nonlinear polaron transport problem which goes beyond the classical description in terms of generalized Langevin equations and thus is able to cover both the low- and high-field regimes.

### C. Eikonal expansion

Similar to the corresponding approximation in scattering theory, the basic idea is that the average motion of the center of mass is characterized by a fixed velocity $v$. It is therefore natural to expand the influence phase in the exponent of the double path integral (19) for the exact collision term to linear order in the fluctuations,

$$\delta\vec{x}(t) = \vec{x}(t) - (\vec{x}_0 + \vec{v}t) , \tag{34}$$

neglecting terms of order $\delta x y^2$. By contrast, the time derivative of the exact influence phase, which may be written as

$$\partial_t i\phi = \frac{2}{\hbar^2}\int_0^t dt' \int \frac{d^3k'}{(2\pi)^3}|\lambda_{k'}|^2 \left\{ \sin\frac{\vec{k}'\cdot\vec{y}}{2} e^{i\vec{k}'[\vec{x}-\vec{x}(t')]} \left[ \cos\frac{\vec{k}'\cdot\vec{y}(t')}{2} \,\mathrm{Im}S(t-t') - \sin\frac{\vec{k}'\cdot\vec{y}(t')}{2} \,\mathrm{Re}S(t-t') \right] \right\} + \{\vec{k}' \to -\vec{k}'\} , \tag{35}$$

is kept in its full generality. Using a partial integration of the kinetic energy term in the form

$$\int_0^t dt_1 \dot{\vec{x}}\cdot\dot{\vec{y}} = (\vec{x}\cdot\dot{\vec{y}} - \vec{x}_0\cdot\dot{\vec{y}}_0) - \int_0^t dt_1 \vec{x}\cdot\ddot{\vec{y}}$$

and

$$\int d^3x \, \exp i\left[ \frac{M}{\hbar}\dot{\vec{y}}_t + \vec{k}' \right]\vec{x} = \left[ \frac{2\pi\hbar}{M} \right]^3 \delta\left[ \dot{\vec{y}}_t + \frac{\hbar\vec{k}'}{M} \right] ,$$

it is straightforward to show that the resulting momentum balance (20) can be written as

$$-\vec{F} = \frac{2}{\hbar^2}\int d^3x_0 d^3y_0 \rho(\vec{x}_0,\vec{y}_0) \int_0^t dt' \int \frac{d^3k'}{(2\pi)^3}|\lambda_{k'}|^2 \int d^3p \, \vec{p} \int \frac{d^3y}{M^3} e^{-i\vec{p}\cdot\vec{y}/\hbar} \times \left\{ e^{i[(M/\hbar)\vec{v}(\vec{y}-\vec{y}_0)+\vec{k}'\cdot\vec{v}(t-t')]} \sin\frac{\vec{k}'\cdot\vec{y}}{2} \int_{\vec{y}_0}^{\vec{y}} D^3y \, A[\vec{y}] \delta\left[ \dot{\vec{y}}_t + \frac{\hbar\vec{k}'}{M} \right] \right. \times \left. \int_0^{\delta\vec{x}(t)} D^3\delta x \exp\left[ -\frac{i}{h} M \int_0^t dt_1 \vec{z}[\vec{y}]\cdot\delta\vec{x}(t_1) \right] \right\} + \{\vec{k}' \to -\vec{k}'\} . \tag{36}$$

Here we have defined functionals of the off-diagonal path $\vec{y}(t_1)$ by

$$A[\vec{y}] = \exp\left\{ \frac{i}{\hbar}\vec{F}\int_0^t dt_1 \vec{y}(t_1) + i\phi[\vec{x}=\vec{v}t,\vec{y}] \right\} \left[ \cos\frac{\vec{k}'\cdot\vec{y}(t')}{2} \,\mathrm{Im}S(t-t') - \sin\frac{\vec{k}'\cdot\vec{y}(t')}{2} \,\mathrm{Re}S(t-t') \right] \tag{37}$$

and

$$\vec{z}[\vec{y}] = \ddot{\vec{y}}(t_1) + \frac{\hbar\vec{k}'}{M}\delta(t_1-t') + \vec{\gamma}[\vec{y}] , \tag{38}$$

with

$$\vec{\gamma}[\vec{y}] = -\frac{4}{\hbar M}\int \frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k} \left\{ \int_0^{t_1} dt_2 \cos\vec{k}\cdot\vec{v}(t_1-t_2) \sin\frac{\vec{k}\cdot\vec{y}(t_1)}{2} \cos\frac{\vec{k}\cdot\vec{y}(t_2)}{2} \,\mathrm{Im}S(t_1-t_2) + \int_{t_1}^t dt_2 \cos\vec{k}\cdot\vec{v}(t_1-t_2) \sin\frac{\vec{k}\cdot\vec{y}(t_2)}{2} \cos\frac{\vec{k}\cdot\vec{y}(t_1)}{2} \,\mathrm{Im}S(t_1-t_2) \right\} . \tag{39}$$

Obviously, the path integral $D^3\delta x$ is now trivial, giving a product of $\delta$ functions, $\prod_{j=1}^{N-1}\delta(\vec{z}_j)$, of the discretized variables $\vec{z}_j$. This in turn fixes the off-diagonal path $\vec{y}(t_1)$, which must obey the equation of motion $\vec{z}[\vec{y}]=0$, i.e.,

$$\ddot{\vec{y}}(t_1)+\frac{\hbar\vec{k}'}{M}\delta(t_1-t')+\vec{\gamma}[\vec{y}]=0 \ . \tag{40}$$

In addition, the integrals over $\vec{p}$ and the final value $\vec{y}=\vec{y}(t)$ may be expressed in the form

$$\int d^3p\,\vec{p}\int\frac{d^3y}{(2\pi\hbar)^3}e^{-i\vec{p}\cdot\vec{y}/\hbar}G(\vec{y})=-i\hbar\frac{\partial G(\vec{y})}{\partial\vec{y}}\bigg|_{\vec{y}=0} .$$

From

$$\sin\frac{\vec{k}'\cdot\vec{y}}{2}e^{i[(M/\hbar)\vec{v}(\vec{y}-\vec{y}_0)+\vec{k}'\cdot\vec{v}(t-t')]}+\{\vec{k}'\to-\vec{k}'\} \ ,$$

this gives a contribution

$$\hbar\vec{k}'\sin[\vec{k}'\cdot\vec{v}(t-t')-M\vec{v}\cdot\vec{y}_0/\hbar] \ ,$$

while all the remaining quantities may be evaluated at $\vec{y}=0$. Therefore, in order to calculate the collision term explicitly, we need to solve the equation of motion (40) with final conditions $\vec{y}(t)=0$ and $\dot{\vec{y}}_t=-\hbar\vec{k}'/M$. Moreover, it is necessary to evaluate the functional determinant of the transformation from the variables $\vec{y}_j$, which appear in $D^3y$ to the $\vec{z}_j$. However, because of the nonlinearity of $\vec{\gamma}[\vec{y}]$, this is not possible in general. To proceed, two further approximations beyond the eikonal expansion have to be made

(a) Take

$$\vec{\gamma}[\vec{y}]\approx-\gamma\dot{\vec{y}} \ , \tag{41}$$

with $\gamma=\gamma(v)$ the collision rate calculated in second-order perturbation theory.

(b) Assume

$$\frac{i}{\hbar}\vec{F}\int dt_1\vec{y}(t_1)+i\phi_1[\vec{x}=\vec{v}t,\vec{y}]\approx0 \tag{42}$$

in $A[\vec{y}]$. In order to justify approximation (b), we expand $\phi_1[\vec{x}=\vec{v}t,\vec{y}]$ to linear order in $\vec{y}$, which gives $i\phi_1\approx-(i/\hbar)\vec{F}_B(v)\int dt_1\vec{y}(t_1)$, with $\vec{F}_B(v)$ the frictional force defined in (30). Since this will cancel the external force $\vec{F}$ on average, assumption (b) is valid at least for large velocities. As a result, there is no *explicit* dependence of the collision term on $\vec{F}$ and thus the field only enters implicitly through $v$. It is important to point out that the expansion in $\vec{y}$ which is valid for large $v$ and is equivalent to the classical limit is only made in the exponent. By contrast, no approximation is made in the $\partial_t i\phi$ term, which is indeed crucial for obtaining the correct quantum-mechanical behavior in the low-field regime at least for small coupling. To see the origin of the approximation (a), we assume that $\vec{y}(t_1)$ is slowly varying in time. Then, with $\vec{y}(t_1)-\vec{y}(t_2)/(t_1-t_2)\approx\dot{\vec{y}}$, we find

$$\vec{\gamma}[\vec{y}]=\frac{2}{\hbar}\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k}\cos\vec{k}\cdot\vec{y}(t_1)\int_0^\infty d\tau\{\sin\vec{k}[\vec{v}+\dot{\vec{y}}(t_1)/2]\tau-\sin\vec{k}[\vec{v}-\dot{\vec{y}}(t_1)/2]\tau\}\,\mathrm{Im}S(\tau)/M \ . \tag{43}$$

Working again to lowest order in $\vec{y}$, we may replace $\cos\vec{k}\cdot\vec{y}(t_1)$ by 1. We then obtain

$$M\vec{\gamma}[\vec{y}]\approx\vec{F}_B(v-\dot{y}/2)-\vec{F}_B(v+\dot{y}/2) \ . \tag{44}$$

Using Eq. (31), this indeed reduces to (41), with

$$\gamma(v)=\frac{\lambda^2\omega_0}{2\pi\hbar Mv^3}\ln(v/\bar{v})\,\theta(v-\bar{v}) \ , \tag{45}$$

provided that $\dot{y}\ll v$. As will be shown below, this effective damping constant $\gamma$ for large velocities is identical with that obtained from a second-order perturbation calculation of the momentum balance. While (45) vanishes for $v<\bar{v}$, we expect that fluctuations in the velocity, which are negligible for large $v$, will lead to a finite damping for all velocities. Therefore we use the approximation (41) quite generally, but take $\gamma(v)=F^{(2)}(v)/Mv$ as the corresponding result obtained from a golden rule calculation; see (58) below. For large $v$ the resulting $\gamma$ then coincides with (45).

With this approximation (40) can be solved explicitly using the ansatz $\vec{y}(t_1)=(\hbar\vec{k}'/M)f(t-t_1)$. The corresponding equation for $f(s)=f(t-t_1)$,

$$\ddot{f}(s)+\gamma\dot{f}(s)+\delta(s-\tau)=0 \ , \quad \tau=(t-t') \ , \tag{46}$$

with initial conditions $f(0)=0$ and $\dot{f}(0)=1$, is solved by

$$f_\tau(s)=\frac{1}{\gamma}[1-e^{-\gamma s}]-\frac{1}{\gamma}\theta(s-\tau)[1-e^{-\gamma(s-\tau)}] \ . \tag{47}$$

Moreover, the functional determinant of the transformation $y_j\to z_j$ is given by $f_\tau(t)$ such that

$$\int_{\vec{y}_0}^0 D^3y\ A[\vec{y}]\delta\left[\dot{\vec{y}}_t+\frac{\hbar\vec{k}'}{M}\right]\int_0^{\delta\vec{x}(t)}D^3\delta x\exp\left[-\frac{i}{\hbar}M\int_0^t dt_1\vec{z}[\vec{y}]\cdot\delta\vec{x}(t_1)\right]$$
$$=\left[\frac{M}{2\pi\hbar}\right]^3[f_\tau(t)]^{-3}\delta\left[\dot{\vec{y}}_t+\frac{\hbar\vec{k}'}{M}\right]\cdot A[\vec{y}] \ , \tag{48}$$

where

$$A[\vec{y}]\approx\left[\cos\frac{\vec{k}'\cdot\vec{y}(t')}{2}\,\mathrm{Im}S(t-t')-\sin\frac{\vec{k}'\cdot\vec{y}(t')}{2}\,\mathrm{Re}S(t-t')\right]e^{-\phi_2[\vec{x}=\vec{v}t,\vec{y}]} \tag{49}$$

is now evaluated at the trajectory $\vec{y}(t')$ defined by (47). Using $\vec{y}_0=-\dot{\vec{y}}_t f_\tau(t)$, the $\delta$ function may be expressed as

$$[f_\tau(t)]^{-3}\delta\left[\dot{\vec{y}}_t+\frac{\hbar\vec{k}'}{M}\right]=\delta\left[\vec{y}_0-\frac{\hbar\vec{k}'}{M}f_\tau(t)\right]. \tag{50}$$

Now for any $\gamma\neq 0$ the fact that $f_\tau(t)$ vanishes exponentially with $t$ implies that $\vec{y}_0(t\to\infty)=0$. Because of the normalization

$$\int d^3x_0\rho(\vec{x}_0,\vec{y}_0=0)=1\ , \tag{51}$$

which holds for an *arbitrary* initial density matrix, we have thus shown explicitly that the asymptotic value of the collision term is independent of the precise initial condition as expected on physical grounds. We mention that for this argument it is important to keep the coupling finite also in the influence phase $i\phi$ appearing in the exponent of (19). Indeed, in a naive perturbation theory in $\lambda$ to second order, which neglects $i\phi$ because the factor $\partial_t i\phi$ is already proportional to $\lambda^2$, the initial condition will *not* be forgotten. The limits $t\to\infty$ and $\lambda\to 0$ are therefore not interchangeable, a problem which will be discussed in more detail in the Appendix.

The final equation for the momentum balance in the steady state then reads

$$\vec{F}=-\frac{2}{\hbar}\int_0^t d\tau\int\frac{d^3k'}{(2\pi)^3}|\lambda_{k'}|^2\vec{k}'\sin(\vec{k}'\cdot\vec{v}\tau)e^{-\phi_2[\vec{x}=\vec{v}t,\vec{y}]}\left[\cos\frac{\vec{k}'\cdot\vec{y}(t-\tau)}{2}\,\mathrm{Im}S(\tau)-\sin\frac{\vec{k}'\cdot\vec{y}(t-\tau)}{2}\,\mathrm{Re}S(\tau)\right]. \tag{52}$$

For the numerical calculation, it is convenient to introduce dimensionless variables $K=k/k_0$, with $k_0=(2M\omega_0/\hbar)^{1/2}$, $z=\omega_0\tau$, and $\tilde{\gamma}=\gamma/\omega_0$. Moreover, we define a dimensionless velocity

$$v_0=\frac{v}{v_t}\ , \tag{53}$$

with $v_t=(2\hbar\omega_0/M)^{1/2}$ the threshold velocity, where the kinetic energy of a free electron is equal to the energy of an optical phonon. Consistent with our expansion to lowest order in $\vec{y}$ of the influence phase in the *exponent* of (19), we evaluate $\phi_2[\vec{x}=\vec{v}t,\vec{y}]$ approximately by using $\sin\vec{k}\cdot\vec{y}/2\approx\vec{k}\cdot\vec{y}/2$ in (23). Performing two time and one angular integration, we then obtain, in dimensionless units,

$$\phi_2[\vec{x}=\vec{v}t,\vec{y}]=\frac{4\alpha}{\pi}K'^2\coth\frac{\beta\hbar\omega_0}{2}\int_{-1}^1 dx[(1-x^2)(1-x'^2)+2x^2x'^2]$$
$$\times\int_0^\Lambda dK\,K^2\frac{\sin^2(2xKv_0-1)z/2}{(2xKv_0-1)^2}\frac{1}{(2xKv_0-1)^2+\tilde{\gamma}^2}\ , \tag{54}$$

where $\Lambda=k_{\max}/k_0$. As pointed out above, $\phi_2$ describes the effect of the fluctuating force due the eliminated phonons. Obviously, this is strongly dependent on the average velocity and vanishes in the limit $v_0\gg 1$. Finally, we introduce a dimensionless external force $\tilde{F}=F/\hbar\omega_0 k_0$. Then in the limit $t\to\infty$ the momentum balance equation (52) can be expressed as

$$\tilde{F}=\frac{2}{\pi}\alpha\int_0^\infty dz\int_0^\Lambda dK'\,K'\int_{-1}^1 dx'\,x'\sin(2x'K'zv_0)\exp(-\phi_2)$$
$$\times\left\{\cos[K'^2d(z)]\sin z+\sin[K'^2d(z)]\coth\left[\frac{\beta\hbar\omega_0}{2}\right]\cos z\right\}, \tag{55}$$

with $d(z)=(1-e^{-\tilde{\gamma}z})/\tilde{\gamma}$ and $\phi_2$ from (55). This is our final result for the nonlinear relation between field and velocity, which will be discussed in detail in the following section.

## III. RESULTS

### A. Weak coupling

In the limit where the interaction may be treated by second-order perturbation theory, we may neglect $\phi_2\sim\alpha$ and replace $d(z)$ by $z$, since the prefactor in (55) is already of order $\alpha$. In terms of the original units, the momentum balance can then be written as

$$\vec{F}^{(2)}=\frac{1}{\hbar}\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k}\int_{-\infty}^{\infty}d\tau\frac{\cos\omega_0(\tau-i\beta\hbar/2)}{\sinh\beta\hbar\omega_0/2}\exp i\left[\frac{\hbar k^2}{2M}-\vec{k}\cdot\vec{v}\right]\tau\ . \tag{56}$$

As was noted by Thornber and Feynman,[5] this result is identical with that obtained from a golden rule calculation,

$$\vec{F}^{(2)}=2\pi\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k}[N_{\rm ph}\delta(\epsilon_{\vec{p}-\hbar\vec{k}}-\epsilon_{\vec{p}}-\hbar\omega_0)+(N_{\rm ph}+1)\delta(\epsilon_{\vec{p}-\hbar\vec{k}}-\epsilon_{\vec{p}}+\hbar\omega_0)] \tag{57}$$

for the loss of momentum of an electron with $\vec{p}=M\vec{v}$ due to phonon absorption and emission. Here $N_{\rm ph}=(\exp\beta\hbar\omega_0-1)^{-1}$ is the equilibrium phonon occupation number at temperature $T$, consistent with our assumption that the lattice remains in equilibrium during the transport process. The evaluation of the integrals in (57) for arbitrary $v$ in the case of a finite Brillouin zone $|\vec{k}|<k_{\rm max}$ is somewhat tedious[10] and will not be presented here. Assuming that $\Lambda=k_{\rm max}/k_0>1$, the resulting dimensionless relation between velocity and force may be written in the form

$$\tilde{F}^{(2)}(v)=\alpha[A(v)N_{\rm ph}+E(v)(N_{\rm ph}+1)]\ . \tag{58}$$

The associated absorption term $A(v)$ is given by

$$A(v)=\begin{cases}\dfrac{1}{v_0}\sqrt{v_0^2+1}-\dfrac{1}{v_0^2}\operatorname{arcsinh}(v_0) & \text{if } v_0\le\dfrac{\Lambda^2-1}{2\Lambda}\ ,\\[2ex] \dfrac{1}{4v_0^2}[\Lambda^2-(v_0-\sqrt{v_0^2+1})^2]-\dfrac{1}{2v_0^2}[\ln\Lambda+\operatorname{arcsinh}(v_0)] & \text{if } v_0\ge\dfrac{\Lambda^2-1}{2\Lambda}\ ,\end{cases} \tag{59}$$

while the emission coefficient $E(v)$ reads

$$E(v)=\begin{cases}\dfrac{1}{v_0}\sqrt{v_0^2-1}+\dfrac{1}{v_0^2}\operatorname{arccosh}(v_0) & \text{if } 1\le v_0\le\dfrac{\Lambda^2+1}{2\Lambda}\ ,\\[2ex] \dfrac{1}{4v_0^2}[\Lambda^2-(v_0-\sqrt{v_0^2-1})^2]+\dfrac{1}{2v_0^2}[\ln\Lambda+\operatorname{arccosh}(v_0)] & \text{if } v_0\ge\dfrac{\Lambda^2+1}{2\Lambda}\ .\end{cases} \tag{60}$$

Clearly, $E(v)\neq0$ only if the velocity is larger than the threshold $v_t$ for single-phonon emission. As may be seen from Fig. 1, where the analytical result (58) is plotted, this threshold leads to a strong increase in the field necessary to produce steady-state velocities larger than $v_t$. In addition, we find that $F^{(2)}(v)$ exhibits a discontinuous derivative at $v_0=(\Lambda^2-1)/2\Lambda$ and $v_0=(\Lambda^2+1)/2\Lambda$. The physical origin of these discontinuities is related to the fact that for $v_0>(\Lambda^2-1)/2\Lambda$ or $v_0>(\Lambda^2+1)/2\Lambda$ energy conservation restricts the angle between $\vec{v}$ and the phonon momentum $\vec{k}$ to a finite range. In particular, for $v_0\gg1$ we need $\vec{k}\cdot\vec{v}\to0$; i.e., phonons can be absorbed or emitted only transversely to the direction of the moving electron. The associated vanishing of the phase space for possible interactions with the lattice leads to $F^{(2)}(v\gg v_t)\to0$ and thus is the basic origin for the instability phenomenon, i.e., $\partial_v F(v)<0$ for $v>v_c$. In fact, this behavior is generic for general electron-phonon interactions of the form (3). For velocities $v\gg v_t$, we have $E=-A=\ln(2v_0)/2v_0^2$ and thus the temperature-dependent factors $N_{\rm ph}$ drop out, giving $F^{(2)}(v\gg v_t)=M\gamma(v)v$ with $\gamma(v)$ as in (45). It is important to point out that for this result it is necessary to keep the maximum phonon wave vector $k_{\rm max}$ finite. Indeed, if $k_{\rm max}$ were set equal to infinity, as is usually done,[7,8] one obtains $E=A=1$ for $v\gg v_t$; i.e., $F^{(2)}(v\gg v_t)$ would approach the finite and temperature-dependent constant $\alpha\coth\beta\hbar\omega_0/2$.

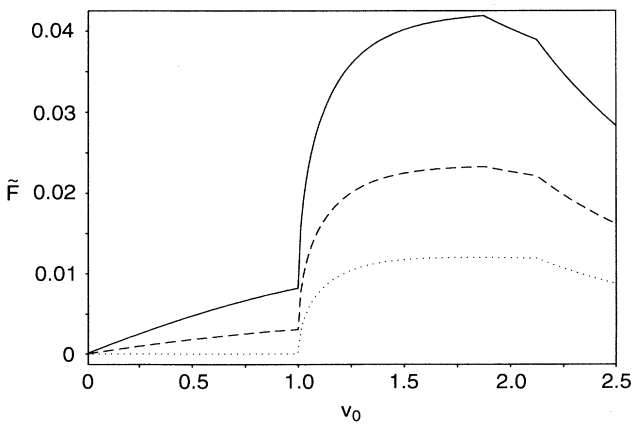


FIG. 1. Dimensionless field $\tilde{F}$ vs normalized velocity $v_0=v/v_t$ obtained from the second-order momentum balance (58) at different temperatures $\beta\hbar\omega_0=0.5$ (solid line), $\beta\hbar\omega_0=1$ (dashed line), and $\beta\hbar\omega_0=10$ (dotted line). The dimensionless cutoff is $\Lambda=4$.

The weak-coupling expression (57) is valid in a strict one-particle description for a *single* electron. In practice, of course, there is a finite electron density with a distribution of velocities $v$. Assuming that the gas of carriers is nondegenerate, this effect may be incorporated in the one-particle description by taking an average over a

Maxwell distribution with mean drift velocity $v$. Starting from the general expression (14) for the one-particle influence phase $\phi$, this is equivalent to the replacement

$$S(t)\rightarrow S(t)e^{-k^2t^2/2\beta M} \tag{61}$$

for the phonon correlation function. Shifting $\tau-i\beta\hbar/2\rightarrow\tau$, the resulting averaged golden rule expression

$$\langle\vec{F}^{(2)}\rangle=\frac{1}{\hbar}\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2\vec{k}\times\int_{-\infty}^{\infty}d\tau\frac{\cos\omega_0\tau}{\sinh\beta\hbar\omega_0/2}\times e^{-i\vec{k}\cdot\vec{v}(\tau+i\beta\hbar/2)}\exp\left[-\frac{\hbar k^2}{2M}\left[\frac{\tau^2}{\beta\hbar}+\frac{\beta\hbar}{4}\right]\right] \tag{62}$$

is then identical with the weak-coupling result obtained by Thornber and Feynman.[5] We note that for arbitrary coupling the replacement (61) affects both the $\partial_t i\phi$ and $i\phi$ terms in the general result (19). Moreover, a decay of the phonon correlation function $S(t)$ would also arise intrinsically from a finite phonon lifetime. In our treatment, however, such effects are neglected since the lattice is assumed to be perfectly harmonic.

In the following we will show that the result (62) is in very good agreement with high-field transport experiments in InSb performed many years ago.[17,18] InSb is a weakly polar semiconductor with Fröhlich constant $\alpha=0.022$, optical phonon energy $\hbar\omega_0=0.0228$ eV, and a small band mass $M=0.0138m_e$.[19] The associated threshold velocity is thus $v_t=7.57\times10^7$ cm/sec, while $F_0=\hbar\omega_0k_0=2.09\times10^4$ eV/cm is the characteristic field. Both experiments were performed at a temperature $T=77$ K, i.e., $\beta\hbar\omega_0=3.45$. The experimental data are shown in Fig. 2 in dimensionless units $\tilde{F}=F/F_0$ versus $v_0=v/v_t$. Here the small dots which go beyond the threshold $v_0=1$ are the results of Glicksman and Steele,[17] while those of Fujisada, Kataoka, and Beer[18] at two different carrier concentrations are denoted by larger open or closed dots, respectively. It is evident that the theoretical result (62), which is the solid line in Fig. 2, gives a very good description of these experiments over more than two orders of magnitude in field or velocity. Empirically, the linear response regime where $F(v)\sim v$ is valid up to about $v\approx0.1v_t$. It should be stressed that there is no fitting procedure involved here since all necessary material parameters are taken from experiment. The good agreement between experiment and theory shows that the dominant source for scattering in InSb at high fields is indeed optical phonons. For comparison, we have also shown the strict one-particle result (58) as a dotted line. It exhibits a very sharp threshold at $v=v_t$, which is washed out in the averaged expression (62). In addition, we have indicated the result obtained for the linear mobility using the Kubo formula[20] as a straight line between both curves. This mobility differs from the one obtained from (62) by a well-known factor $3k_BT/2\hbar\omega_0$.[1] Evidently, the measurements are strongly in favor of the Thornber-Feynman result, which is also obtained here. As will be shown in the Appendix, this gives indeed the correct mobility in a situation where the steady-state distribution is spread over all momenta.

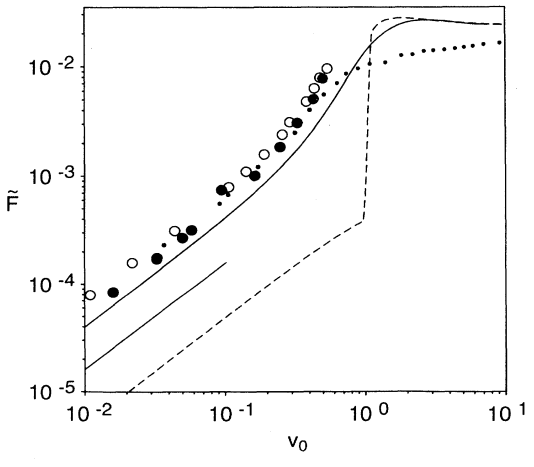


FIG. 2. Dimensionless-field–velocity relation (solid line) obtained from (62) in comparison with the experimental results on InSb at 77 K (Refs. 17 and 18). The dashed line is the strict one-particle result (58). The Kubo result (A5) for the linear mobility is indicated by the straight line.

### B. Strong coupling

For strong interaction between electrons and optical phonons, perturbation theory is not applicable and thus the full momentum balance (55) has to be evaluated numerically. In order to compare our results with those of Thornber and Feynman,[5] we have chosen $\alpha=3$. The resulting dimensionless relation $\tilde{F}(v_0)$ in the vicinity of the threshold $v_0=1$ and at $\beta\hbar\omega_0=5$ is shown in Fig. 3 together with the corresponding Thornber-Feynman result. Obviously, the current-voltage characteristics are very different, both quantitatively and qualitatively. It is only at unphysically large velocities $v\gg v_t$—which cannot be

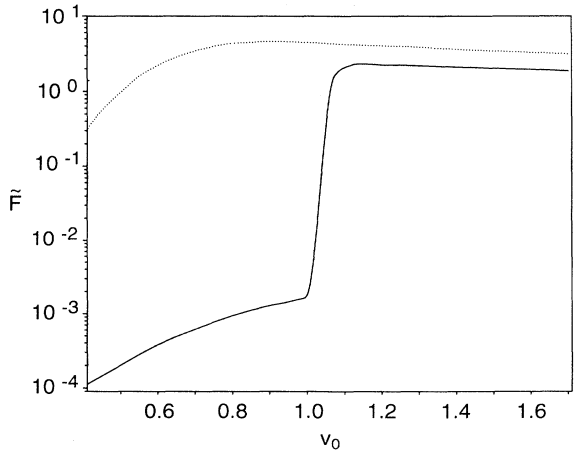


FIG. 3. Dimensionless-field–velocity relation for $\alpha=3$ and $\beta\hbar\omega_0=\Lambda=5$. The dotted line is the corresponding Thornber-Feynman result.

seen on the scale of our figure—that both methods give identical results. In particular, our method predicts a rather sharp threshold around the bare value $v=v_t$, while in the Thornber-Feynman theory this is strongly washed out and also shifted to lower velocities. The reason for this is that Thornber and Feynman have used the equilibrium effective polaron mass $M^*$, which is considerably larger than $M$ and thus leads to a reduced threshold velocity $v_t \sim M^{-1/2}$. In our present theory such an effect does not occur. Indeed, we believe that at velocities of order $v_t$ the phonon polarization cloud, which is the physical origin of the mass enhancement in equilibrium, is stripped off and the electron's inertia is essentially that of a bare particle. This conclusion remains unchanged if we include the averaging via (61), which increases somewhat the necessary fields for a given velocity, and, in contrast to the weak coupling case, does not wash out the threshold as long as $\beta\hbar\omega_0$ is much larger than 1. As a second point, it is striking that the Thornber-Feynman method predicts maximal fields $F_c$ at low temperatures which are about twice as large as those obtained here.

A comparison between theory and experiment for systems with large coupling constants $\alpha$ is difficult since it is hard to generate free carriers in such systems and to avoid heating effects in strong fields. Thornber and Feynman have compared their predictions qualitatively with experiments on $Al_2O_3$where $\alpha \approx 2.7$. They give an estimate for the maximum field $F_c$ which is correct in order of magnitude; however, because of considerable uncertainties in the parameter values, a quantitative comparison is difficult. In particular, there are no detailed velocity-field curves for such systems. We have thus chosen indirect measurements of the drift velocity of carriers in AgCl which are estimated from the Hall mobility in weak magnetic fields.[21] The experimental results for the drift velocity (and the photocurrent $Q_x$, open dots) are shown in Fig. 4. They exhibit a sharp increase in field around a certain threshold velocity. The horizontal line in the figure, which is denoted by $\frac{1}{2}v_{\rm op}$, is the average velocity expected for the so-called "ideal streaming motion."[21] This is based on the simple picture that electrons are freely accelerated from $v=0$ up to $v=v_t$ where they emit one optical phonon and thus are back at $v=0$. If the time interval between successive accelerations and phonon emission is shorter than all other scattering times, the associated average velocity is just $v_t/2$. Obviously, this is a highly oversimplified model and indeed our results show that the actual threshold where $F(v)$ starts to increase strongly is near $v \approx v_t$. Noting that the velocity scale in Fig. 4 is logarithmic, this is in fact consistent with the experimental data. In any case the measurements show a flat plateau in $v(F)$ in agreement with our very steep $F(v)$ relation (see Fig. 3). By contrast, in the theory of Thornber and Feynman, $v(F)$ starts to level off much below $v_t$ and does not exhibit a flat region. As a second point, we estimate the characteristic field $F_0=\hbar\omega_0 k_0$, which is about $1.2\times10^5$ eV/cm. From Fig. 3 the field at the beginning of the sharp rise is approximately $2\times10^{-3}F_0=240$ eV/cm, which also roughly agrees

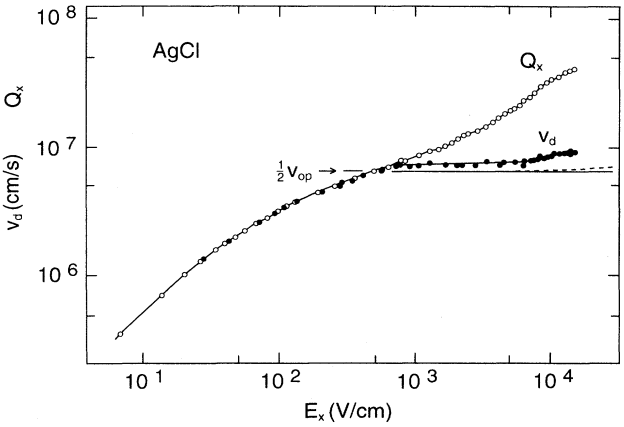


FIG. 4. Measurements of the drift velocity vs field (solid dots) and the photocurrent (open dots) in AgCl taken from Ref. 21.

with the measured value (the difference between $\alpha=3$ and the estimated value $\alpha \approx 2$ for AgCl as well as that between $\beta\hbar\omega_0=5$ and the much larger experimental value $\beta\hbar\omega_0 \approx 60$ is rather small around $v \approx v_t$; see also Thornber and Feynman).[5] We therefore believe that the experiments indeed support our results, but clearly a more detailed comparison is required to draw more definite conclusions.

## IV. DISCUSSION

In this paper we have presented a method to calculate the nonlinear mobility of polarons with arbitrary strength of the electron-phonon coupling. Our main objective was to reexamine the Feynman path-integral formulation for nonlinear transport, avoiding the application of the equilibrium variational principle. Our method is based on an eikonal expansion of the exact collision term of the Wigner distribution function. For weak coupling our results coincide with those of Thornber and Feynman. However, for strong coupling there are large differences, in particular regarding the value and sharpness of the threshold velocity $v_t$ as well as the magnitude of the maximum attainable field $F_c$, which allows a steady state with optical phonon scattering only. Our prediction that the phonon-induced mass renormalization is no longer relevant at velocities of order $v_t$ is supported by measurements of high-field transport in AgCl.

A crucial feature of our approach is that it includes the velocity-dependent fluctuations around the average trajectory. Such fluctuations have been discussed widely in the context of the generalized Langevin equation approach.[14] Indeed, the $v$-dependent and anisotropic diffusion constants $D_{\parallel}(v)$ and $D_{\perp}(v)$ introduced in (32) and (33) are effectively equivalent to an imaginary contribution $-iDk^2$ to the phonon frequencies.[14] The resulting "collision broadening" effects lead to a reduction of the mobility obtained from Eq. (29).[14] As was explained in Sec. II B, however, the Langevin equation treats the electron classically.[22] In general, this is valid only for

large fields or velocities. Qualitatively, the classical limit may also be justified for large coupling $\alpha \gg 1$, since $\exp\{-\phi_2[\vec{x},\vec{y}]\}$ then strongly suppresses the offdiagonal elements $\vec{y}$.[11] It is difficult, however, to make this argument more precise. In the present approach, we go beyond the classical description by keeping the full quantum-mechanical expression for the time derivative of the influence phase $\partial_t i\phi$ in the exact collision term (19). An alternative quantum treatment of nonlinear polaron transport was proposed several years ago by Su, Chen, and Ting.[23] It is based on the Keldysh technique, which is formally equivalent to the Feynman-Vernon influence functional method. Using a Gaussian approximation for the velocity fluctuations, they derive an energy balance equation which leads to $F(v)$ curves in close agreement with those of Thornber and Feynman. In view of our present quite different results, this is surprising since the basic approach to the problem appears to be similar. At present, we do not understand the precise origin of these differences. Further investigations and in particular more detailed comparison with experiments are therefore necessary.

## ACKNOWLEDGMENT

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## APPENDIX

Here we want to discuss the problem of the "correct" linear mobility $\mu_l = \lim_{v\to 0} v/F$ of a Fröhlich polaron to lowest order in the coupling constant $\alpha$. We start from the weak-coupling result (62), which is the drifted Maxwellian average of the single-particle expression (56). Splitting this into an emission and an absorption contribution and taking the momentum cutoff $k_{\max}\to\infty$, an expansion to first order in $v$ gives the inverse linear mobilities as

$$\mu_{e,a}^{-1} = \frac{2}{3}\alpha M\omega_0 \frac{\beta\hbar\omega_0}{2\sinh\beta\hbar\omega_0/2}\left[\frac{\beta\hbar\omega_0}{\pi}\right]^{1/2} \times\left[K_1\left[\frac{\beta\hbar\omega_0}{2}\right]\pm K_0\left[\frac{\beta\hbar\omega_0}{2}\right]\right], \tag{A1}$$

where $K_{0,1}(z)$ are modified Bessel functions. At low temperatures $T \ll \hbar\omega_0/k_B = T_0$, we find that

$$\mu_e^{-1}(T\ll T_0) = \tfrac{2}{3}\beta\hbar\omega_0 2\alpha M\omega_0 N_{\rm ph}\ , \tag{A2}$$

while

$$\mu_a^{-1}(T\ll T_0) = \tfrac{2}{3}\alpha M\omega_0 N_{\rm ph}\ . \tag{A3}$$

Obviously, the absorption contribution is negligible compared with the emission term, which therefore determines the *full* inverse mobility. By contrast, at high temperatures $T \gg T_0$, emission and absorption contribute equally with

$$\mu_e^{-1}(T\gg T_0) = \mu_a^{-1}(T\gg T_0) = \tfrac{4}{3}\alpha M\omega_0\left[\frac{T}{\pi T_0}\right]^{1/2}. \tag{A4}$$

In the following we will concentrate on the low-temperature regime $T \ll T_0$. There it turns out that the absorption contribution (A3) is identical with the *total* inverse mobility, which is obtained from a strict single-particle calculation with no averaging. Indeed, in the linear response limit $v_0\to 0$, we obtain from (58) that $\tilde{F}(v_0) = \frac{2}{3}\alpha N_{\rm ph} v_0$ since phonon emission does not contribute. A similar result is obtained by evaluating the exact quantum-mechanical Kubo-formula for $\mu_l$ perturbatively in $\alpha$, which gives[20]

$$\mu_l^{\rm Kubo} = (2\alpha M\omega_0 N_{\rm ph})^{-1}\ . \tag{A5}$$

It differs from the single-particle or the averaged absorption contribution to $\mu^{-1}$ by a factor of 3. In the literature $\mu^{\rm Kubo}$ is usually called $\mu^{\rm RTA}$ because the identical expression follows from a relaxation time approximation of the classical Boltzmann equation.[1] Since the Kubo formula gives an *exact* expression for any linear transport coefficient, however, we prefer to call it $\mu^{\rm Kubo}$. Evidently, the Thornber-Feynman and Kubo results (A2) and (A5) differ by a factor $3k_BT/2\hbar\omega_0$. The reason for this discrepancy is usually attributed to the problem in which order the limits $\lambda\to 0$ and $t\to\infty$ are taken.[1,4] As has been shown explicitly in Sec. II C, this is in fact a subtle point since taking $\lim\lambda\to 0$ *first,* the initial condition will not be forgotten in principle. In the following it will be shown that the conflicting results (A2) and (A5) describe different physical situations. In particular, we will see that the disagreement between both expressions is due to the different way in which thermal averaging enters in the calculation of either the *inverse* mobility $\mu^{-1}$ or the mobility $\mu$ itself. As a result, phonon *emission* does not contribute to $\mu_l^{\rm Kubo}$ as $T\to 0$;[1] however, it does contribute—or is even dominant—in the Thornber-Feynman theory. To see this it is instructive to derive the result (A2) for the total inverse mobility by calculating $\mu^{-1}$ from a force-force correlation function to second order in $\lambda$.[24,25] Introducing the force operator

$$\hat{F}_z = -iV^{-1/2}\sum_k \lambda_k k_z \exp(i\vec{k}\cdot\vec{q})x_k\ , \tag{A6}$$

we have

$$\mu_l^{-1} = \lim_{\epsilon\to 0}\int_0^\infty dt\, e^{-\epsilon t}\int_0^\beta d\alpha\langle \hat{F}_z(t)\hat{F}_z(t=i\hbar\alpha)\rangle\ , \tag{A7}$$

with $\hat{F}_z(t)$ the usual Heisenberg time evolution. Since $\hat{F}_z$ is already of order $\lambda$, the expectation value may be calculated with the uncoupled Hamiltonian $H_0$. The associated density matrix is therefore

$$\rho_0 = \frac{e^{-\beta H_B}}{Z_B}\left[\frac{\beta}{2\pi M}\right]^{3/2} e^{-\beta\vec{p}^2/2M}\ . \tag{A8}$$

It is then straightforward to show that

$$\mu_l^{-1}=\pi\beta\hbar\int\frac{d^3k}{(2\pi)^3}|\lambda_k|^2k_z^2\int d^3p\left[\frac{\beta}{2\pi M}\right]^{3/2}e^{-\beta\vec{p}^2/2M}$$
$$\times[N_{\rm ph}\delta(\epsilon_{\vec{p}-\hbar\vec{k}}-\epsilon_{\vec{p}}-\hbar\omega_0)+(N_{\rm ph}+1)\delta(\epsilon_{\vec{p}-\hbar\vec{k}}-\epsilon_{\vec{p}}+\hbar\omega_0)]\ . \quad \text{(A9)}$$

Evaluating this we find that $\mu_e^{-1}=\mu_a^{-1}$ at *all* temperatures. The resulting total inverse mobility is identical with (A1) (i.e., the $\pm K_0$ contributions are absent here). It is now evident that the Thornber-Feynman result is obtained by thermally averaging the inverse mobility $\mu^{-1}$. By contrast, in the Kubo formulation one averages $\mu$. As a result, only absorption contributes to $\mu^{\rm Kubo}$, while both absorption and emission enter in (A9). The absence of emission contributions in linear response is usually argued to indicate that $\mu^{\rm Kubo}$ is the correct result.[1] In practice, however, the actual nonequilibrium distribution function for a finite density of electrons may lead to a different situation. Indeed, Thornber and Feynman assume that this is a drifted Maxwellian, which may be established, for instance, by electron-electron scattering. If this is the case, there is a steady population of electrons with sufficiently large momenta such that phonon emission is possible even for a very small average drift velocity. In such a situation—which appears to be realized experimentally (see Sec. III A)—the result (A2) of Thornber and Feynman gives the "correct" linear mobility.


[1]See, e.g., F. M. Peeters and J. T. Devreese, in *Solid State Physics,* edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1984), Vol. 38, p. 81.
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