Effect of the polaron-induced nonparabolicity of the energy-momentum relation on the dynamics of transport electrons

P. Warmenbol and F. M. Peeters
Department of Physics, University of Antwerp (Universitaire Instelling Antwerpen), Universiteitsplein 1, B-2610 Wilrijk (Antwerpen), Belgium

J. T. Devreese
Department of Physics, University of Antwerp (Universitaire Instelling Antwerpen), Universiteitsplein 1, B-2610 Wilrijk, (Antwerpen), Belgium;
University of Antwerp (Rijksuniversitair Centrum Antwerpen), Groenenborgerlaan 171, B-2020 Antwerpen, Belgium;
Eindhoven University of Technology, NL-5600 MB Eindhoven, The Netherlands
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The influence of the electron-phonon-interaction—induced nonparabolicity in the electron energy-momentum relation $E(k)$ on the electron transport in crossed electric and magnetic fields is investigated within a simple Shockley-type model. Different models for the polaron nonparabolic energy-momentum relation are considered. Numerical results are given for AgCl and are compared with the results for a parabolic $E(k)$ relation. The Hall factor and the magnitude of the drift velocity are sensitive to this polaron effect, while the Hall angle is almost insensitive.

I. INTRODUCTION

The electron-phonon interaction influences the static and dynamical properties of an electron moving in a polar semiconductor or in an ionic crystal. The changes are more pronounced for materials with a large electron-phonon Fröhlich coupling constant α, e.g., they are more important for AgCl ($\alpha=1.84$) than for InSb ($\alpha=0.02$). The influence of the electron-phonon interaction on the properties of the electron are twofold:[1] (i) the interaction with virtual phonons leads to a mass renormalization, a binding energy, and a nonparabolic energy-momentum $[E(k)]$ relation, and (ii) the interaction with real phonons is responsible for the dissipation of the electron energy. The latter effect will be treated within the Shockley model which assumes that the electron emits instantaneously a LO phonon if its energy equals the energy of a LO phonon. This model describes approximately the carrier streaming motion in polar semiconductors.

Several works have been devoted to the study of the energy-versus-momentum relation of polarons.[2–10] Theories ranging from effective-mass approximations,[2,3] over perturbation theory[5,10] (up to fourth order), to variational methods[7] have been developed. The purpose of the present paper is to investigate the effect of this polaron $E(k)$ relation on the dynamics of polaron transport, i.e., on experimentally accessible quantities like the drift velocity, the Hall factor, and the mobility. To get a qualitative idea of the influence of this nonparabolicity on the electron transport properties, we will make the following simplifying assumptions: (i) scattering with acoustical phonons and impurities will be neglected and only the emission of LO phonons will be retained, and (ii) the emission process of LO phonons is described using the Shockley model.

The above assumptions lead to qualitatively meaningful results for high electric fields, low temperature, and pure samples. The aim of the present calculation is to get the right order of magnitude and the correct qualitative trend of the influence of the electron-phonon-interaction—induced nonparabolicity on the transport properties of electrons in crossed electric and magnetic fields. The present rather simple calculation may motivate a more elaborate study based on the Boltzmann equation (e.g., by performing a Monte Carlo simulation).

In a recent paper by Brazis *et al.*,[11] the effect of a nonparabolic conduction band on the electron heating was investigated. There, a different type of nonparabolicity was considered, namely, the band nonparabolicity (e.g., the $\mathbf{k}\cdot\mathbf{p}$ interaction) which is not a consequence of the electron-phonon interaction. For example, in InSb this band nonparabolicity can be fairly accurately described by the Kane model, while the polaron contribution to the nonparabolic $E(k)$ relation is very small in this material. The situation is different in the strongly ionic materials AgCl and AgBr, where the intrinsic conduction band is practically parabolic, but where the electron-phonon interactions can induce appreciable nonparabolic effects in the $E(k)$ relation. It is the latter situation which is of interest in the present paper.

In Sec. II the $E(k)$ and $v(k)$ relations are discussed briefly, following four different approaches. In Sec. III the solution of the polaron equations of motion in crossed electric and magnetic fields is studied for the different $E(k)$ relations. The field dependence of the drift velocity, the tangent of the Hall angle, and the Hall factor are analyzed in Sec. IV, adopting the classical transport model of Komiyama, Masumi, and Kajita (KMK) of Ref. 12, which is an extension of the Shockley model to the case of crossed fields. Motivated by the recent experimental work of KMK (Refs. 12 and 13) on the silver halides, we will apply our numerical calculations to the case of AgCl, which has a fairly large electron-phonon coupling constant, i.e., $\alpha=1.84$, and where we expect the effects under investigation will show up in a pronounced way.

II. POLARON CORRECTIONS TO THE ELECTRON ENERGY-MOMENTUM RELATION

Of the various investigations of the electron energy-momentum relation found in the literature, only a few will be considered, all of which have a different physical content.

(i) A *free electron model with a bare band mass* m_b, assuming no effect of the electron—LO-phonon interaction. The dispersion relation is then given by $E_1(k)=k^2$, where the energy is in units of the LO-phonon energy $\hbar\omega_{LO}$ and the momentum is in units k_{LO}, which is defined by $k_{LO}^2/2m_b=\hbar\omega_{LO}$.

(ii) A *quasiparticle model with an effective mass* m^*, assuming the mass renormalization to be momentum independent, $E_2(k)=k^2m_b/m^*$, where $m^*/m_b=1/(1-\alpha/6)$ is the polaron mass renormalization, which is valid for $\alpha \ll 6$.

(iii) Considering the electron LO-phonon interaction as a small perturbation, "*improved Wigner-Brillouin perturbation theory*" (IWBPT),[14] which is equivalent to the *one-phonon Tamm-Dancoff approximation*,[5] gives to second order

$$E_3(k)=k^2-\Delta E \ , \tag{1a}$$

where

$$\Delta E=\begin{cases}\dfrac{\alpha}{k}\arcsin\left[\dfrac{k}{\sqrt{1-\alpha-\Delta E}}\right], & k<k_{crit}\\[2ex] \alpha\dfrac{\pi}{2}, & k>k_{crit}\ ,\end{cases} \tag{1b}$$

and the wave vector k_{crit} is defined by

$$\varepsilon_3(k_{crit})=E_3(k_{crit})-E_3(0)=1\ . \tag{1c}$$

For example, for $\alpha=0.07$ we obtained $k_{crit}=1.02$ and for $\alpha=1.84$, $k_{crit}=1.23$.

(iv) *The variational ansatz of Larsen* (VAR) in Ref. 7 is a combination of the one-phonon Tamm-Dancoff approximation[5] and the Lee-Low-Pines transformation.[3] Larsen attempts to extend the range of validity to intermediate coupling strengths. This gives

$$E_4(k)=k^2+\left[1-\frac{E_4(k)-k^2}{2k^2}\right]F(k,E_4(k),\alpha)\ , \tag{2}$$

with

$$F=\frac{4\pi\alpha}{(2\pi)^3}\times\int d\mathbf{q}\frac{(2\mathbf{q}\cdot\mathbf{k})^2}{q^2(1+q^2)^2[E_4(k)+\alpha-k^2-1+2\mathbf{q}\cdot\mathbf{k}-q^2]}\ .$$

Both IWBPT and VAR have the property

$$\lim_{\varepsilon(k)\to 1^-}\frac{\partial E}{\partial k}=0\ , \tag{3}$$

as conjectured by Schultz[4] and Whitfield and Puff.[6] In Larsen's theory k_{crit}, defined by $\varepsilon_4(k_{crit})=E_4(k_{crit})-E_4(0)=1$, is found to be $k_{crit}=1.02$ for $\alpha=0.07$ and $k_{crit}=1.43$ for $\alpha=1.84$. As a consequence of Eq. (3), the quasiparticle polaron velocity $v(k)=\partial E(k)/\partial k$ approaches zero in the limit $\varepsilon(k)\to 1^-$.

Figure 1 shows the real part of the polaron energy-momentum relation as obtained from these four approaches. For $\alpha=0.07$ (GaAs) the four approaches do not differ much from each other because α is small compared to 1 (IWBPT and VAR give the same numerical results up to two to three digits for this value of α). When $\alpha=1.84$ [AgCl (Ref. 15)] (see Fig. 1) the different physical approaches behind the four theories result in qualitative as well as quantitative differences for $E(k)$. IWBPT and VAR clearly show a bend-over in the region $\varepsilon(k)\approx 1$, as expressed by Eq. (3).

Figure 2 shows the velocity versus momentum for the same approaches as considered above. VAR and IWBPT give similar qualitative results. We expect that for $\alpha=1.84$, VAR is more accurate than IWBPT. Whether or not the behavior of VAR and IWBPT for $k>k_{LO}$ is physical (or observable) is open to discussion. Therefore, we also included a different $E(k)$ relation [denoted by Rayleigh-Schrödinger perturbation theory (RSE)], which for $k \ll k_{LO}$ coincides with VAR up to first order in α, but which has a continuous derivative around $k\approx k_{crit}$. The starting point is the $E(k)$ relation as obtained within Rayleigh-Schrödinger perturbation theory, which can be

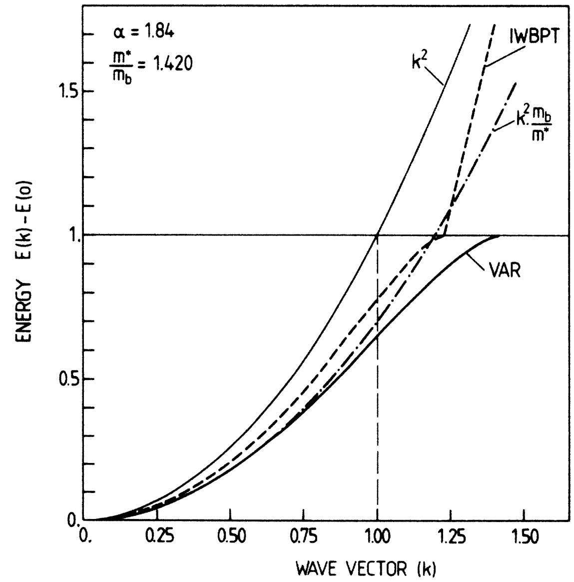

FIG. 1. Real part of the energy versus momentum according to different theories for the electron—LO-phonon coupling constant $\alpha=1.84$ (AgCl).

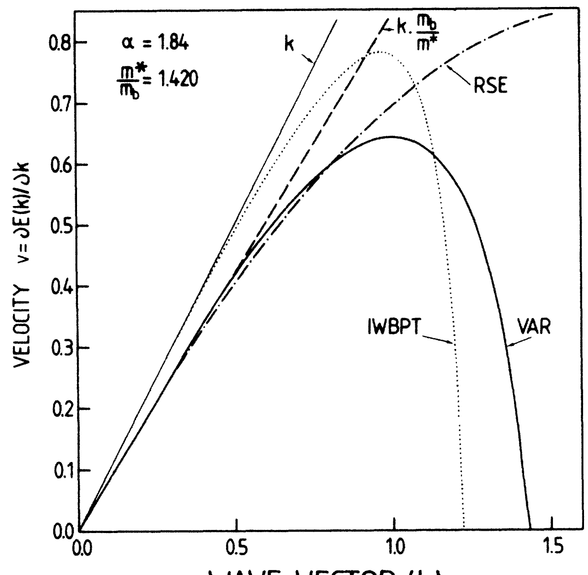

FIG. 2. Wave-vector dependence of the polaron velocity as obtained from the following different theories: (i) free electron with bare band mass (thin solid curve), (ii) free electron with effective mass (dashed curve), (iii) expansion of the Rayleigh-Schrödinger perturbation theory (dashed-dotted curve), (iv) variational calculation of Larsen (VAR) (Ref. 2) (thick solid curve), and (v) IWBPT (dotted curve).

expanded for small momentum:

$$\begin{aligned} E_5(k) &= k^2 - \frac{\alpha}{k}\arcsin(k) \\ &\simeq k^2 - \frac{\alpha}{k}\left(k + \tfrac{1}{6}k^3 + \tfrac{3}{40}k + \cdots\right) \\ &\simeq E_5(0) + \frac{k^2}{2m^*(k)} , \end{aligned} \tag{4}$$

with

$$m^*(k) = m^*(0)(1+\delta k^2)$$

and

$$\delta \simeq 3\alpha \Big/ \left[80 \left[1 - \frac{\alpha}{6} \right] \right] .$$

The velocity becomes

$$v(k) = \frac{\partial E(k)}{\partial k} = \frac{k}{(1+\delta k^2)^2} . \tag{5}$$

The resulting expression (4) exhibits a nonparabolic relation between E and k and is only valid for $k \ll k_{\rm LO}$. We will apply Eq. (4) over the whole k range in order to investigate the importance of the bend-over in the $E(k)$ result around $k \sim k_{\rm LO}$ for VAR and IWBPT. The RSE curve in Fig. 2 is for $\delta = 0.10$ ($\alpha = 1.84$). As a consequence of Eq. (3), VAR and IWBPT give a velocity, which tends to zero as the energy approaches the LO-phonon energy. The three other theories give a monotonously increasing velocity as a function of wave vector. The nonlinearity of the velocity-wave-vector relation is most pronounced for VAR and IWBPT, less so for RSE. The existence of polarons, which have large wave vector but low velocity, is expected to modify the picture of free-electron transport.

III. MOTION OF POLARONS IN CROSSED ELECTRIC AND MAGNETIC FIELDS

The classical equation of motion for a charge carrier in crossed electric $\mathbf{E}(E,0,0)$ and magnetic field $\mathbf{B}(0,0,B)$ is

$$\hbar \dot{\mathbf{k}}(t) = e\mathbf{E} + \frac{e}{m}[\mathbf{v}(\mathbf{k}) \times \mathbf{B}] , \tag{6}$$

where the velocity $\mathbf{v}(\mathbf{k})$ depends on the $E(k)$ relation one assumes. Solution of Eq. (6) is standard for the parabolic case.[12] It consists of circular orbits around the center $\mathbf{c}(0,-(1/\zeta),k_z)$ with $\zeta = v_{\rm LO}(B/E)$ and $mv_{\rm LO}^2/2 = \hbar\omega_{\rm LO}$. For the nonparabolic case, one is forced to solve Eq. (6) numerically because $E(k)$ is not known analytically.

Figure 3 shows the solutions of the equation of motion [Eqs. (6)] with the initial condition $(k_x(t=0), k_y(t=0)) = (0,0)$ for (i) the parabolic case with effective mass m^* (PAR) and (ii) for Larsen's variational calculation (VAR). From Fig. 3 it is apparent that VAR gives trajectories which for small k are indistinguishable from PAR but which for $k > k_{\rm LO}$ tend to become parallel to the electric field direction; the reason being that the velocity, and consequently also the Lorentz force, fall off to zero as k approaches $k_{\rm crit}$. For $\zeta > 2$, the trajectories are closed orbits which differ little from PAR.

IV. CLASSICAL TRANSPORT FOR POLARONS IN CROSSED ELECTRIC AND MAGNETIC FIELDS

The model of KMK (Ref. 12) at zero temperature is generalized here to the case of a general $E(k)$ relation. As in the Shockley[16] model we assume that the probabili-

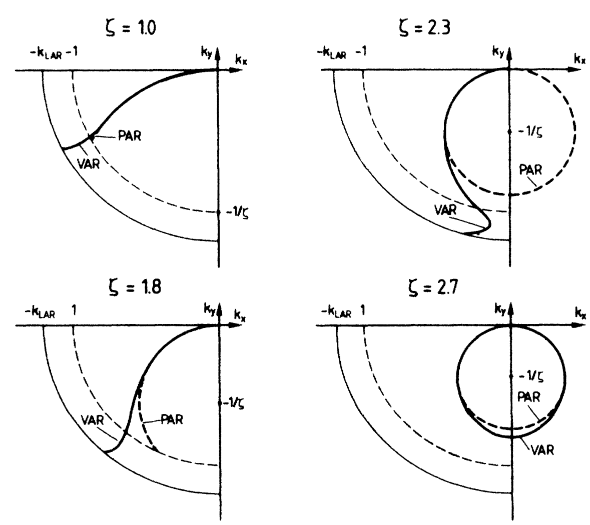

FIG. 3. Solution of the electron equation of motion in k space for the following two different $E(k)$ relations: (i) free electron with effective mass (PAR) (thick dashed curve) and (ii) variational calculation of Larsen (VAR) (thick solid curve), and for different values of the dimensionless field.

ty for LO-phonon emission is 1 at the polaron circle $\varepsilon(k)=1$ and the electron is scattered back towards the ground state, i.e., towards $k=0$. As in Ref. 12, we define a time interval $T_{\rm op}(\zeta)$ by

$$\varepsilon(k_{\rm crit})=E(k(t=T_{\rm op}(\zeta)))-E(0)=\hbar\omega_{\rm LO}\ , \tag{7}$$

which is the time needed by the electron to gain an energy $\hbar\omega_{\rm LO}$ when it starts from the origin $k=0$. The average drift-velocity components are defined by the time average

$$v_d^{x,y}(\zeta)=\frac{1}{T_{\rm op}}\int_0^{T_{\rm op}} v_{x,y}(\zeta,t)dt\ . \tag{8}$$

The Hall factor r_H and the tangent of the Hall angle are, respectively, defined by

$$r_H(\zeta)=\frac{1}{\zeta}\frac{v_d^y(\zeta)}{v_d^{x2}(\zeta)} \tag{9}$$

and

$$\tan\theta_H(\zeta)=\frac{v_d^y(\zeta)}{v_d^x(\zeta)}\ . \tag{10}$$

Figure 4 shows the drift-velocity components v_d^x and v_d^y and the Hall factor r_H as a function of ζ for three different $E(k)$ relations: (i) effective-mass theory, (ii) RSE, and (iii) VAR, for an electron-phonon coupling constant $\alpha=1.84$ (AgCl). Qualitatively the three quantities v_d^x, v_d^y, and r_H behave similarly for the three different theories with increasing ζ. The validity of the KMK model is questionable in the region where ζ is close to 2, where accumulation of the electrons on closed orbits becomes very important. Therefore, we consider only $\zeta<2$. For $\zeta<1$ the velocity v_d^x is significantly lowered due to the nonparabolicity effect. The lowering is 16% for VAR compared to PAR at $\zeta=1$ but only 5% for RSE. The same general trend holds for v_d^y. As a consequence, $r_H(\zeta)$ tends to higher values [see Eq. (9)] for $\zeta<1$ by some 13% for VAR and only 2% for RSE compared to PAR. It is apparent from Fig. 4 that RSE and VAR show the same trend but differ quantitatively. This difference can again be attributed to the conjectured Eq. (3) or, equivalently, the behavior of the velocity and the trajectories in the region $k\geq k_{\rm LO}$ (see Figs. 2 and 3). Thus, the bend-over of the $E(k)$ relation at $k=k_{\rm crit}$ leads to appreciable quantitative differences in v_d^x, v_d^y, and r_H, but no qualitative differences are observed.

Figure 5 shows the tangent of the Hall angle as a function of ζ for the same $E(k)$ models as in Fig. 4. In addition, experimental results from KMK (Ref. 12) are shown for sample M1CB at a fixed electric field of 3.3 kV/cm. Because the external fields enter only through $\zeta\sim B/E$ in the KMK model, the present theoretical results are independent of the absolute values of these fields. Experimentally there is considerable spread only if $\zeta>1$ and if the absolute value of the external fields is varied. It is apparent from Fig. 5 that the three theories show the same qualitative behavior and differ only slightly from each other in the range $1<\zeta<2$. This can be explained by observing that (i) $\tan\theta_H$ is a relative quantity [see Eq. (10)] and (ii) both v_d^x and v_d^y are lowered by the same relative amount.

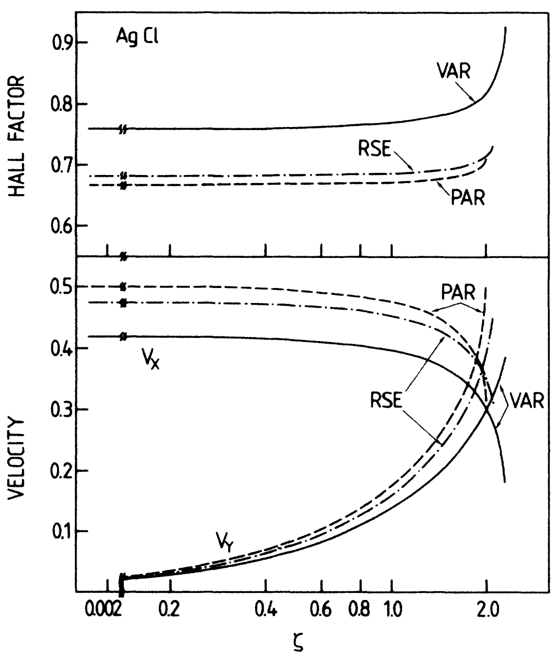

FIG. 4. Field dependence of velocity components v_d^x and v_d^y and Hall factor r_H as obtained from the KMK model [Eqs. (8) and (9)] for $\alpha=1.84$ (AgCl) and for the following different $E(k)$ relations: (i) free electron with effective mass (PAR) (dashed curve), (ii) expansion of the Rayleigh-Schrödinger perturbation theory (dashed-dotted curve), and (iii) variational calculation of Larsen (VAR) (solid curve). Velocity in units of $v_{\rm LO}$, where $v_{\rm LO}^2/2m_b=\hbar\omega_{\rm LO}$ with $\hbar\omega_{\rm LO}$ the LO-phonon energy.

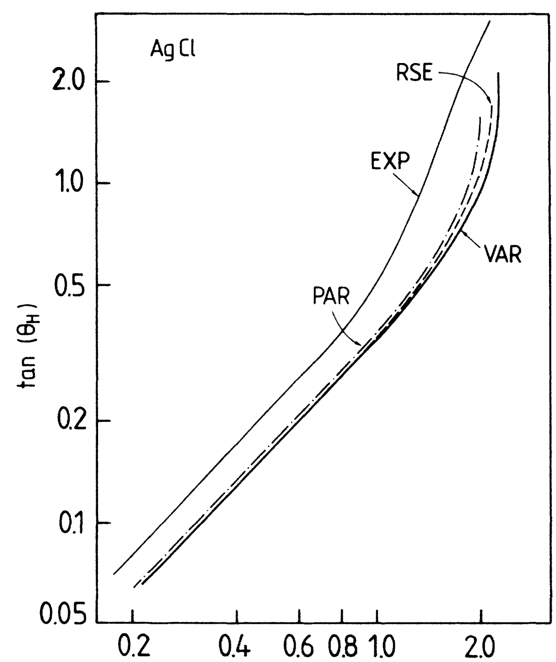

FIG. 5. Field dependence of the tangent of the Hall angle for $\alpha=1.84$ and for the same $E(k)$ relations as in Fig. 3. Also indicated are experimental values from Komiyama *et al.* for AgCl for sample M1CB at a fixed electric field of 3.3 kV/cm.

It may be surprising that the effects of nonparabolicity on the electron dynamics are appreciable for *all* values of ζ. This can be understood as follows: in the present model the polaron performs a streaming motion when $\zeta<2$. During one acceleration cycle the polaron obtains all energies between 0 and $\hbar\omega_{\rm LO}$, irrespective of the value of ζ. Changing ζ only alters the time the polaron needs to perform one cycle. For large values of ζ the present model is clearly not valid because, e.g., impurity scattering, acoustical phonon scattering, ... will force part of the electrons in a nonstreaming state, which is not taken into account in the present calculation. As a consequence, the practical applicability of the present results for ζ around 2 is questionable.

V. DISCUSSION

The present investigation allows us to draw the following qualitative conclusions on the influence of the nonparabolicity of the $E(k)$ relation on the polaron motion in crossed electric and magnetic fields.

(i) The nonparabolicity changes appreciably the form of the momentum distribution function in the region $1<k<k_{\rm crit}$ for values of ζ close to 1 (see Fig. 3).

(ii) The nonparabolicity raises the threshold value $\zeta_{\rm crit}$, for which closed orbits exist in k space, to values $\zeta_{\rm crit}>1$.

(iii) The nonparabolicity lowers the absolute value of the drift-velocity components over the whole region of $\zeta<2$.

(iv) The nonparabolicity increases the Hall factor r_H over the whole $\zeta<2$ region.

(v) The nonparabolicity does *not* have an appreciable effect on $\tan\theta_H$.

The experimental field dependence of $\tan\theta_H$ for AgCl at high electric fields (a few kV/cm) differs significantly from the theoretical description (i.e., Monte Carlo simulation,[17] KMK model,[12] ...) in the region $1<\zeta<2$. We have shown that due to the fact that $\tan\theta_H$ is rather insensitive to the nonparabolicity of $E(k)$, this discrepancy between theory and experiment cannot be explained by an electron-phonon-induced nonparabolic $E(k)$ relation when the LO-phonon emission process is treated within the simple Shockley model. Because of the simplicity of the model, the present calculation does not exclude the possibility that the nonparabolicity is the origin of the discrepancy between theory and experiment. Nevertheless, the present results should rather be considered as a plausible qualitative argument against the suggestion of Komiyama in Ref. 13 that nonparabolicity can explain the discrepancy.

Intuitively we would expect that mobility experiments on the strongly ionic crystal AgCl, such as were done by Komiyama *et al.*,[12] should be able to show clearly the effect of polaron nonparabolicity. The reason why the experiments of Ref. 12 do not give any confirmation for this nonparabolicity is that they measured the Hall mobility μ_H ($\mu_H=\tan\theta_H/B$), which is almost insensitive to this polaron nonparabolicity. This is the underlying reason why in Ref. 17 we were able to explain the low magnetic field data of Ref. 12 with the simple parabolic energy-momentum relation. The present study shows that only a direct measurement of the drift velocity and/or the Hall factor is able to give evidence for this polaron-induced nonparabolicity.

There is yet, as far as we know, no *direct* evidence for the existence of electron-phonon-induced nonparabolicity in the energy-momentum relation of polar semiconductors and ionic crystals. Theories obeying Eq. (5) predict a reasonably strong effect on the magnitude of the drift velocity. The validity of conjecture [Eq. (5)] can in this way be checked by a direct measurement of the drift velocity via, e.g., a time-of-flight experiment.

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