Drift velocity for acoustic polarons in 1*D* conductors

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The drift velocity $v_0(E)$ is calculated for an acoustic polaron with strong coupling in a 1*D* polymer chain. The limiting cases of low and high temperatures are analyzed. The functional dependence $v_0(E)$ exhibits a characteristic saturation in strong fields near the sound velocity *s*. This behavior of the drift velocity was recently observed in polydiacetylene, PDA [K. J. Donovan and E. G. Wilson, J. Phys. C **18**, L51 (1985); Philos. Mag. **B44**, 9, 31 (1981)].

The majority current carriers in 1*D* polymer chains are polarons[1,2] which are formed as a result of a strong interaction of electrons with 1*D* acoustic phonons.[3] The deformation interaction which is linear in the phonon coordinates Q_k is described in the system of units with $m = \hbar = s = 1$ by the Hamiltonian

$$H = \sum_k Q_k\Big(V_k e^{ikx} + \frac{1}{2}k^2 Q_{-k}\Big) - \frac{1}{2}\frac{\partial^2}{\partial x^2}, \qquad V_k = Dk(2M_1)^{-1/2} = k(\alpha\omega_1)^{1/2}. \tag{1}$$

Here m is the electron mass, s is the sound velocity, D is the strain energy, M_1 is the mass of the primitive cell, and $\omega_1 = 2s/a$ (a is the lattice constant). In the case of polydiacetylene, $(CH)_X$, the dimensionless constant of the strong coupling, $\alpha = D^2/2M_1\omega_1 \gg 1$ is 4 ($D = 3$ eV, $s = 10^6$ cm/s, $a = 1.4$ Å, $M_1 = 13$ a.u.; Ref. 4), while in the case of polydiacetylene, PDA, it is 12 ($D = 3.7$ eV, $s = 3.6\times 10^5$ cm/s, $a = 4.9$ Å, $M_1 = 420$ a.u.; Ref. 5).

The spectrum $\varepsilon(p)$ for a strong-coupling acoustic polaron with a velocity $v \sim 1$ is determined by the classical minimum of H in a coordinate system which is moving at a velocity v (Ref. 6):

$$\epsilon = J + pv, \quad p = M^* v, \quad J = J_1 + J_2, \tag{2}$$

where J_1, J_2, and M^* are determined by the wave function of the ground state, $\psi_0(x)$, for an electron in a polaron well:

$$2J_1 = \int dx(\partial\psi_0/\partial x)^2, \;\; 2J_2 = w(v^2-1)^{-1}, \;\; M^* = w(1-v^2)^{-2}, \;\; w = 2\alpha\int dx\, \psi_0^4(x). \tag{3}$$

The electron levels in the well $\tilde\varepsilon_n$, and the electron wave functions $\psi_n(\xi)$ are described by the following expressions[7–13] in dimensionless units introduced in terms of the renormalized coupling constant $\tilde\alpha = \alpha(1-v^2)^{-1} = 1$:

$$\psi_0(\xi) = (\sqrt{2}\cosh\xi)^{-1}, \quad \tilde\epsilon_0 = -1, \quad \psi_{\tilde q}(\xi) = e^{i\tilde q\xi}(\tanh\xi - i\tilde q)/(1-i\tilde q), \quad \tilde\epsilon_{\tilde q} = \tilde q^2. \tag{4}$$

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Here $\psi_0(\xi)$ is the Rashba-Holstein[7,8] ground-state wave function, the $\psi_{\tilde{q}}(\xi)$ are the wave functions of the continuous spectrum, where $\xi = x/\tilde{\alpha}$, and the energy $\varepsilon_{\tilde{q}}$ is expressed in units of $\alpha^2 ms^2$.

The quantities[9] $J = -\tilde{\alpha}^2/6$ and $M^* = -2\partial J/\partial v^2$ determine the spectrum of a polaron $\varepsilon(p)$, and its asymptotic behavior at large momentum, $p \gg \alpha^2$:

$$\epsilon(p) \approx p\left(1 - \frac{3}{4}(2\alpha^2/3p)^{1/3}\right), \quad v(p) \approx 1 - \frac{1}{2}(2\alpha^2/3p)^{1/3} \tag{5}$$

Consequently, the asymptotic value $v = 1$ in the limit $p \to \infty$ is reached far more slowly in this case than for a piezopolaron, for example.[6]

To calculate the mobility μ, we use the general method of Volovik and Édel'shtein[14] for a piezoelectric polaron with strong coupling. This method has also been used to calculate the mobility of an ordinary polaron.[14,15] Another method for deriving equations for μ in the case of an ordinary polaron was proposed by Mel'nikov and Volovik.[16] At $v \sim 1$ we transform to a coordinate system moving at a velocity v, thereby introducing a Doppler shift of the phonon frequencies, $\omega_k = |k| \to \tilde{\omega}_k = \omega_k - kv$. We then apply the general formalism of Ref. 14, based on Bogolyubov-Tyablikov[17] and Lee-Low-Pines[18] transformations. At phonon-reservoir temperatures $T \ll \alpha^2$, the mobility μ is described by the Fokker-Planck equation

$$eE\,\partial f/\partial p = \frac{\partial}{\partial p}\left(Af + B(\partial f/\partial p)\right), \; A = Bv/T. \tag{6}$$

The coefficient B is expressed in terms of the two-phonon scattering amplitude $W_{kk'}$ and the Planck phonon occupation numbers N_k (Refs. 14–16):

$$B = \pi \sum_{k,k'} N_k(N_{k'} + 1)|W_{kk'}|^2 (k - k')^2 \delta(\tilde{\omega}_k - \tilde{\omega}_{k'}), \; \tilde{\omega}_k = \omega_k - kv. \tag{7}$$

The quantities $W_{kk'}$ satisfy an equation for the scattering of phonons by a heavy particle[14–16] with a Doppler shift, (7):

$$W_{kk'} = V_{kk'} - \sum_{k''} V_{kk''} D_{k'k''} W_{k''k'}, \; D_{kk'} = 2\tilde{\omega}_{k'}/(\tilde{\omega}_k^2 - \tilde{\omega}_{k'}^2 + i\delta), \tag{8}$$

$$(\tilde{\omega}_k \tilde{\omega}_{-k'})^{1/2} V_{kk'} = \sum_q v_{0q}(k) v_{q0}(-k')(\epsilon_0 - \epsilon_q)^{-1}, \; v_{0q}(k) = V_k \int dx\, \psi_0^*(x) e^{ikx} \psi_q(x)$$

$$= \frac{i\pi 2^{-1/2} \tilde{k} V_k}{(1 - i\tilde{q})\,\mathrm{ch}\left(\frac{\pi}{2}(\tilde{k} + \tilde{q})\right)}. \tag{9}$$

The field satisfies $E \sim E_0 \sim \alpha^2/el \ll \alpha^2/e\tilde{a}$ where $\tilde{a} \sim \alpha^{-1}$ is the size of the polaron, and $l \sim T^2/Bv$ is its mean free path. The field therefore does not perturb an electron in a well; it simply accelerates the polaron to velocities $v \sim 1$, without causing its destruction. A higher polaron energy $\varepsilon \sim \alpha^2 \gg v/l$ improves the condition for the applicability of the kinetic equation.[6] The condition $T \ll \alpha^2$ ensures that the thermal phonon momenta are small, $k \sim T \ll p \sim \varepsilon$; this condition is necessary for the validity of the Fokker-Planck expansion.[6]

At low temperatures $T \ll \alpha$, the equation of Ref. 8 can be solved by the general method of Ref. 14, through an expansion in $k,k' \sim T \ll k'' \sim \alpha$ and through the use of the well-known identities[14,17] for $V_{kk'}$. The final expressions for $W_{kk'}$ and B are

$$W_{kk'} = V_k V_{-k'} kk'(M^*)^{-1} (\tilde{\omega}_k \tilde{\omega}_{-k'})^{-3/2}, \quad B = B_0(T) A_0 \varphi_0(v), \quad B_0(T) = T^5/\alpha^2, \tag{10}$$

$$A_0 = \frac{3}{5}(2\pi)^3, \quad \varphi_0(v) = \varphi(v) + \varphi(-v), \quad \varphi(v) = \frac{15}{\pi^4}(1+v)^3 \chi_0 \int_0^\infty \frac{k^2 dk \exp(k(\chi_0 - 1))}{\sinh k \sinh k\chi_0}, \tag{11}$$

$$\chi_0 = \frac{1-v}{1+v}.$$

The general solution $f(p)$ for the Fokker-Planck equation in (6) and the expression for the drift velocity $v_0(E)$ are

$$\ln f(p) = -T^{-1}\epsilon(p) + eE\int_0^p dp' B(p'), \quad v_0 = C_0^{-1}\int dp v(p) f(p), \quad C_0 = \int dp f(p). \tag{12}$$

It follows that at $T \ll \varepsilon$ and $E \sim E_0 = A_0 B_0(T)/eT$ the velocity v_0 is determined by the extremum of $\ln f(p)$ and satisfies the equation

$$v_0 \varphi_0(v_0) = \tilde{E}, \quad \tilde{E} = E/E_0, \quad E_0 = A_0 B_0(T)/eT. \tag{13}$$

Figure 1 is a plot of the functional dependence $v_0(\tilde{E})$. At $\tilde{E} < 1$, the function $v_0(\tilde{E})$ is nearly linear and determines the mobility $\mu = s/E_0$, while at $\tilde{E} \gtrsim 1$ the quantity $v_0(\tilde{E}) = 1$ is constant. This result follows from expression (12) for v_0, in which a divergence of the integrals over p occurs as $p \to \infty$. In real systems, the divergence is cut off near the Brillouin momentum $k_0 \sim a^{-1} \gg \alpha^2$, so that we have $v_0(\tilde{E}) \sim v(k_0) < 1$ [see (5)], and this value cannot reach 1. At $E \gg 1$, the momentum is $p \approx k_0$, and the system is no longer described by the continuum model; accordingly, our results are only qualitative in this field region. At $\tilde{E} \gg 1$ the polaron begins to sense the finite width of the band, and the velocity $v_0(\tilde{E})$ may begin to decrease.[19]

At higher temperatures, $\alpha \ll T \ll \alpha^2$, the integral term in (8) is small, proportional to the parameter $\alpha/T \ll 1$, so that we have $W_{kk'} = V_{kk'}$. In this case we have $B_0(T) = T^2\alpha, A_0 = 1{,}14, \varphi_0(v) = (1-v)$ at $(1-v) \gtrsim \alpha/T$. It follows from Eqs. (12) and (13) that we have $v_0(\tilde{E}) = 1$ at all $\tilde{E} > \alpha/T = \tilde{E}_1$. Again in this case, the characteristic momenta are $p \approx k_0$, since a divergence occurs at large p in the integrals in

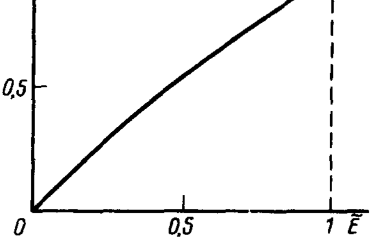

FIG. 1. Drift velocity of an acoustic polaron, v_0, versus the field $\tilde{E} = E/E_0$ at low temperatures.

(12). In this situation we have

$$v_0(\widetilde{E}) \approx v(k_0) < 1 \quad \text{at} \quad \widetilde{E} \gtrsim \widetilde{E}_1 \quad \text{and} \quad (1-v) \gtrsim \alpha/T.$$

This effect has recently been observed by Donovan and Wilson[5] in PDA at room temperature. In their experiments, they observed an approximately constant value $v_0 = 0.715$ over a broad field range, $E \sim 1$–10^4 V/cm. Our estimate of the field, $E_1 \sim s(\alpha ms)^2/e\hbar \sim 10$ V/cm with $\alpha \sim 10$, $s \sim 10^5$ cm/s, and $m \sim 0.1 m_0$ (m_0 is the mass of the free electron),[5] agrees with these results. The value[5] $v_0 = 0.715$, according to expression (5) with $p = k_0$, then leads to the extremely reasonable value $k_0 = 0.8$ of the standard value, π/a.

We note in conclusion that Schüttler and Holstein[12] recently reported some qualitative estimates of the mobility μ in weak fields at low velocities, $v \ll 1$. In weak fields, their estimates agree with our results. Wilson[11] has attempted to calculate the functional dependence $v_0(E)$ in strong fields, but the correct expression for this quantity was not found in Ref. 11 because of several arbitrary assumptions and certain errors.

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