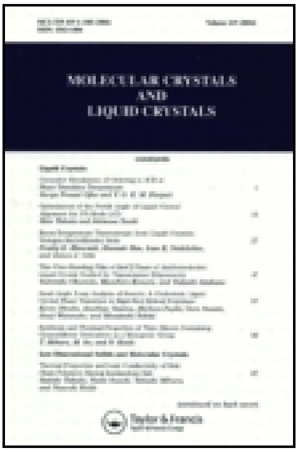

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Free Soliton and Polaron Transport and Photoconductivity in *Trans*-Polyacetylene

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Free soliton transport in transpolyacetylene has been observed through magnetic resonance experiments and in photo-induced absorption and photoconductivity measured within picoseconds after irradiation by above-gap photons. By inference free polaron transport has also been observed in the latter experiments. The transport of these excitations is determined by phonon and defect scattering. The dispersion of these freely propagating excitations and theoretical calculations of their diffusion rate and mobility parallel to the chains, as determined by acoustic phonon scattering, are reviewed. Agreement of the theory with the measured diffusion rate at room temperature is good. Agreement is achieved also at low temperatures, where phonon scattering is ineffective, by introducing barriers of height 0.01 eV. Such barriers are expected for *cis*-inclusions in *trans*-polyacetylene. The calculations are consistent with the observed insensitivity to temperature of the 50 ps photoconductivity, although they eliminate the possibility that the insensitivity is due to the carriers being hot. Experiments are suggested for separating soliton and polaron contributions to photoconductivity and further elucidating the transport of these excitations.

1. INTRODUCTION

The first observation of free soliton transport in *trans*-polyacetylene was through magnetic resonance experiments in undoped material.[1] After considerable controversy there is now wide agreement that these experiments reveal a defect with spin, the neutral soliton, diffusing rapidly along the chains.[2] Deduction from the experimental data of the magnitude of the diffusion constant $D_{\parallel}$ is difficult, however, in that a fraction of the solitons is trapped, that fraction varying with temperature (and with sample), and the trapping has different effects on different types of magnetic resonance measurements. As pointed by Clarke and Scott,[2] the crux of the problem of measuring $D_\parallel$ lies in determining what fraction of the spins is trapped. That has been done with some care by two different experimental groups. Nechtschein *et al.*[3] determined the fraction trapped vs. temperature from ESR measurements and used this in the determination of $D_\parallel$ from proton NMR data. Their results are shown in Figure 1. The results for $D_\parallel$ vs. temperature T of Mizoguchi *et al.*,[4] who studied electron spin resonance, are also presented in Figure 1. Observation of the decay of photo-induced dichroism also gave early evidence for free soliton transport.[5] $D_\parallel$ deduced from this observation for 300 K is in good agreement with that deduced from magnetic resonance.

For the small concentrations usually present the neutral solitons may be thought of as a one-dimensional gas with thermal energy. It is expected that $D_\parallel$ be determined by collisions with phonons and with defects. After discussion of the dispersion relation in section 2a, we will review briefly the calculation of $D_\parallel$, and the mobility $\mu_\parallel$ of

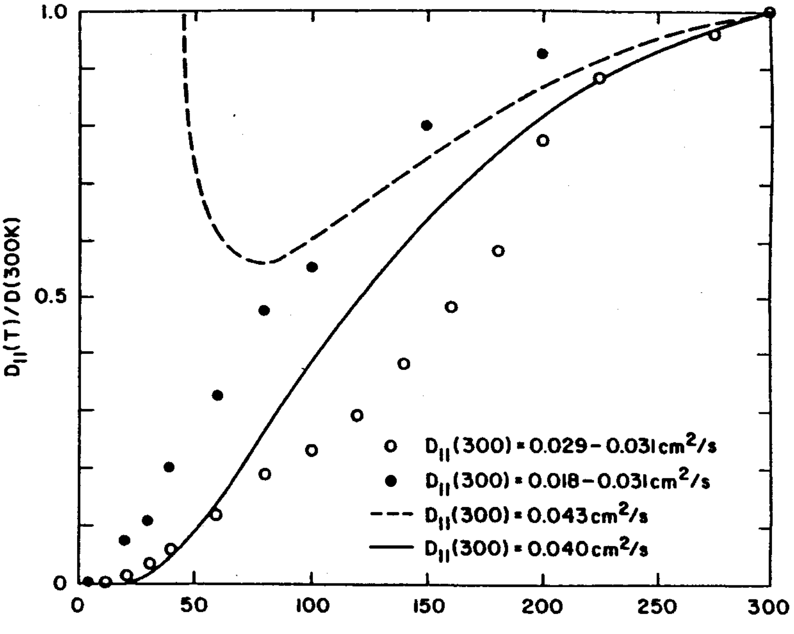

FIGURE 1 Intrachain diffusion constant of neutral solitons in *trans*-$(CH)_x$ normalized to 300 K value. Closed circles: data of Ref. 3; Open circles, data of Ref. 4. Dashed line, the theoretical results for phonon scattering only; solid line, the theoretical results for phonon scattering plus barriers of 0.01 eV.

charged solitons, due to scattering by phonons and defects. Comparison with experiment will also be taken up in section 2. Freely diffusing or drifting polarons should have very similar properties to those of solitons so far as phonon scattering is concerned; their $D_{\parallel}$ and $\mu_{\parallel}$ will also be taken up in section 2.

Evidence for freely moving polarons in *trans*-polyacetylene comes from recent photo-induced absorption measurements at 25 K.[6] In these experiments the charged soliton population was monitored by its characteristic midgap absorption. Irradiation by a 5 ps pulse of above-gap photons produced a large number of charged solitons, which mostly decayed within ~1 ps. However, 20 ps after the pulse was over the charged soliton population was found to rise again, peaking at ~40 ps before it again decayed.[6] The explanation that best fits the data is that some polarons were also generated by the initial pulse and these polarons, diffusing freely, converted neutral solitons present as defects in the sample to charged solitons.

Measurements of photoconductivity within picoseconds[7] or even nanoseconds[8,9] after irradiation by photons with above-gap energy gave much larger photocurrents, with quite different temperature dependence, than those found earlier[10] microseconds and even milliseconds after the irradiating pulse. It was inferred that the short-time photoconductivity represents the transport of freely moving solitons and, on the evidence of the photoinduced absorption experiments,[6] polarons. The long-time photoconductivity has been attributed to electron hopping between trapped charged solitons,[10] presumably those of the earlier photogenerated solitons or converted neutral solitons that have escaped rapid recombination by falling into relatively inaccessible traps. In section 3 we will review the experimental data on picosecond and nanosecond photoconductivity and the inferences therefrom. We also suggest some experiments to help elucidate the detailed properties of freely propagating solitons and polarons.

2. SOLITON AND POLARON DIFFUSION RATES AND MOBILITY

The calculations of diffusion rates and mobility are very similar for soliton and polaron; we will discuss them explicitly for solitons and indicate where modifications are necessary for polarons. The calculations will be carried out for $D_{\parallel}$ and the Einstein relation used to obtain $\mu_{\parallel}$.

2a. Dispersion relation

One piece of information required for the calculation is the relation between the energy E_k and wave vector k of the soliton. For small k or low speed, v

$$E_k \xrightarrow[k\to 0]{} \frac{2\Delta_o}{\pi} + \frac{\hbar^2 k^2}{2m_s^o}, \tag{1}$$

where the low energy soliton mass m_s^o is given by[11]

$$m_s^o = 4Mu_o^2/3b\xi. \tag{2}$$

Here the first term in (1) represents the soliton creation energy,[12] Δ_o being the half-gap, and the second term the kinetic energy.[11] The quantity M is the mass of a C-H group, u_o is the (static) displacement of each C-H to form the dimerized chain from a uniform chain with spacing b between C-H groups, ξ is the half-width of the soliton, given by

$$\xi = 2t_o b/\Delta_o, \tag{3}$$

t_o being the transfer integral. For *trans*-polyacetylene (2) and (3) lead to $m_s^o \simeq 5\, m_e$, where m_e is the free electron mass.[11]

For higher energies the $E_k - k$ relation is complicated by the existence of a maximum velocity c, which arises because the motion of a soliton is accomplished by motion of the C-H groups along the chain. As a result c is of the order of the sound velocity v_s, which has been measured as 1.5×10^6 cm/s.[13,14,15] Numerical calculations give $c = 2.7\, v_s$.[14,15] The existence of a limiting velocity indicates that the soliton mass is a function of its speed v, approaching infinity as $v \to c$. Such a "relativistic" behavior of mass with velocity is familiar for solitons obtained for other one-dimensional systems, *e.g.*, from sine-Gordon and ϕ^4 field theories. The pseudo-relativistic form for the dispersion relation obtained analytically for those cases suggests the generalization for solitons in *trans*-polyacetylene[16]

$$E_k = (\hbar^2 c^2 k^2 + (m_s^o c^2)^2)^{1/2} + \overline{E}, \tag{4}$$

where

$$\overline{E} = (2\Delta_o/\pi) - m_s^o c^2. \tag{5}$$

It is readily seen that the dispersion relation given by (4) and (5) reduces correctly to (1) in the low-velocity limit and gives constant velocity c in the high-velocity limit. It should be noted that for polarons, whose low-velocity mass is only $\sim m_e$, thermal velocities are so large that it is necessary to use the relation (4) rather than (1) above ~100 K.[17]

2b. Phonon scattering

The phonons that can affect the diffusion and drift rates of thermal solitons and polarons at room temperature and below are the acoustic phonons because the energy of the optical phonons in polyacetylene, ~0.2 eV, is much too high. We have calculated the scattering due to interaction of a soliton with a single phonon. Although the phonons and a stationary soliton are both normal modes of the system, the first order matrix element will not vanish for a *moving* soliton.

To determine which phonons are instrumental in scattering solitons or polarons we use the conditions of conservation of momentum and energy. For the case of phonon emission which takes the soliton from k to k', the former condition gives

$$q = k - k' \tag{6}$$

while the latter condition gives, for the case that the initial and final velocities are small compared to c,

$$\hbar v_s q = \frac{\hbar^2 k^2}{2m_s^o} - \frac{\hbar^2 k'^2}{2m_s^o}. \tag{7}$$

Solution of (6) and (7) gives

$$q = 2k - 2m_s^o v_s/\hbar = 2m_s^o(v - v_s). \tag{8}$$

Thus emission is possible only for solitons with $v > v_s$. Equations for q of the form of (8) are obtained also for phonon absorption,[17] there being two because for absorption k' may be either parallel or antiparallel to k.

Significantly, it is found that, for either emission or absorption, the phonons that interact with a soliton or polaron have wavelengths $(=2\pi/q)$ long compared to the length of the soliton or polaron.[16,17] The scattering may then be thought of as due to the compression or expansion of the soliton by the phonon. In that case the matrix element for the scattering process in which $k \rightarrow k'$, and the number of phonons N_q with wavevector q goes to N'_q, may be taken[18,17]

$$M_{k,k'} = \left\langle k', N'_q \left| E_D \frac{\partial u(x)}{\partial x} \right| k, N_q \right\rangle \tag{9}$$

where E_D is the shift in soliton energy per unit strain and $\partial u(x)/\partial x$ the strain due to an acoustic wave propagating in the x (chain) direction with displacement of the point at x given by $u(x)$. E_D may be calculated from the energy shift δE_k due to a uniform strain $\delta b/b$. Neglecting the kinetic energy term in (1) because it is small compared to the creation energy, we obtain

$$E_D = \frac{\delta E_k}{\delta b/b} \rightarrow b\frac{\partial E_k}{\partial b} = b\frac{\partial}{\partial b}\left(\frac{2}{\pi}\Delta_o\right). \tag{10}$$

Taking the b dependence of Δ_o from the gap equation for the dimerized lattice,[12] we find[16]

$$E_D = -(8\alpha b/\pi)(1 + 1/2\lambda)\,[(\exp - (1/2\lambda)] \tag{11}$$

where $\alpha = -\partial t_o/\partial b$,

$$\lambda = 2\alpha^2/\pi k t_o \tag{12}$$

and K is the force constant. With $b = 1.22$ Å, $\alpha = 4.1$ eV/Å,[11] and $\lambda = 0.19$,[12] $E_D = 3.3$ eV for the soliton. For the polaron E_D is larger by a factor $\sqrt{2}$ because the creation energy is larger by that factor.[19]

Evaluation of the matrix element (9) yields[17,20]

$$|M_{k,k'}|^2 = \left(\frac{\hbar}{2MNv_s q}\right) E_D^2 q^2 \left\{ \begin{array}{ll} N(q) & \delta_{k',k+q} \\ (N(q)+1) & \delta_{k',k-q} \end{array} \right. \tag{13}$$

where N is the number of C-H groups in the chain, and the upper factor in braces is for absorption, the lower for emission. The mean free time for scattering by absorption or emission is obtained from the Golden Rule as:[17,20]

$$1/\tau_{\substack{ab\\em}} = (Nb/2\pi) \int dk'(2\pi/\hbar)\, |M_{k,k'}|^2\, \delta(E_{k'} - E_k \mp \hbar\, v_s q) \tag{14}$$

where the upper sign corresponds to phonon absorption, the lower to emission. With E_k given by (4) the integration (14) must be carried out numerically. $D_\parallel$ must then be obtained numerically from

$$D = \langle v^2 \tau \rangle \tag{15}$$

where

$$\frac{1}{\tau} = \frac{1}{\tau_{abs}} + \frac{1}{\tau_{em}}. \tag{16}$$

The mobility is then conveniently calculated from the Einstein relation

$$\mu_\parallel = D_\parallel e / k_B T. \tag{17}$$

2c. Comparison with experiment

The calculated $D_\parallel$ vs. T for phonon scattering is shown as a dashed line in Figure 1. The 300 K value is within ~10 to 30% of the experimental values, which have considerable scatter. Nevertheless, the agreement is impressive because it involves no arbitrary constants. It is seen that the agreement between the experimental $D_\parallel$ and the values calculated for phonon scattering is reasonable down to ~150 K. Below that temperature, however, the theory predicts a sharply increasing $D_\parallel$ because the number of phonons that can scatter decreases sharply, while the experimental $D_\parallel$ continues to decrease. The continuing decrease of $D_\parallel$ is reasonably attributed to defects or trapping. The simplest kind of defect would be a barrier of height V_o, which would essentially cut off the contribution to transport of solitons with $E_k < V_o$. The effect of such a barrier on $D_\parallel$ may be obtained by choosing the lower limit for integration in (15) as V_o instead of 0. It was found that a good fit to the experimental data could be obtained by choosing $V_o = 0.01$ eV, which leads to $D_\parallel$ given by the solid line in Figure 1. The prime candidate for such a barrier, suggested earlier by Gibson *et al.*,[21] is remanent *cis*-linkages in *trans*-polyacetylene, to which they attribute a barrier height ~0.01 eV. Further, they demonstrated by measurements of infrared absorption that there is always a minimum of 5% of *cis*-linkages in *trans*-$(CH)_x$ obtained by the Shirakawa process.

We can use the Einstein relation Eq. (17) to obtain $\mu_\parallel$ vs. T from $D_\parallel$ vs. T. The results are shown in Figure 2. It is seen that $\mu_\parallel \approx 1$ cm^2/Vs at 300 K. The different T-dependence of $\mu_\parallel$ deduced from the

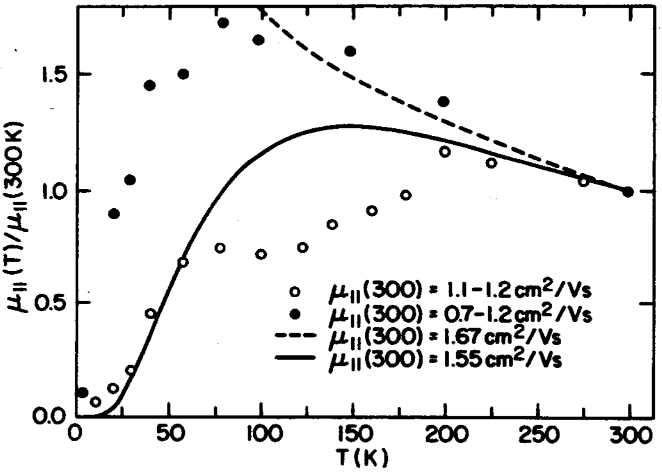

FIGURE 2 Intrachain mobility of solitons in *trans*-$(CH)_x$ (normalized to 300 K value) calculated from $D_{\parallel}e/k_BT$, the $D_{\parallel}$ values taken from Figure 1. Closed circles, $\mu_{\parallel}$ from data of Ref. 3; Open circles, $\mu_{\parallel}$ from data of Ref. 4; Dashed line, theoretical results for phonon scattering only; Solid line, theoretical results for phonon scattering plus barriers of 0.01 eV.

two different sets of experimental data could be due to genuine differences in trapping between the different samples.

$\mu_{\parallel}$ vs. T calculated as described above for polarons scattered by phonons is shown in Figure 3. Because the correct value of c is unknown, the calculations are shown for different c values. No attempt has been made to include defect scattering. It is quite possible that the polaron will be less scattered by conjugation defects, such as *cis*-inclusions, because its passage does not require reversal of the dimerization pattern of the chain.

3. SHORT-TIME PHOTOCONDUCTIVITY

As indicated earlier, measurements of photoconductivity within 50 ps after a pulse of above-bandgap photons gave very large photocurrents, corresponding to a conductivity of 0.3 ohm^{-1} cm^{-1}.[7] With the average 300 K $\mu_{\parallel}$ for solitons and polarons 1 to 2 cm²/Vs, σ of 0.3 ohm^{-1} cm^{-1} corresponds to 10^{18} carriers/cm³. As pointed out by Sinclair *et al.*, this number is consistent with the observations of the decay of photo-induced absorption. If every initial photon had created a pair of solitons, as is reasonably assumed,[22] the initial number of carriers/cm³ would be over 10^{20}/cm³; in 50 ps, however, the observed

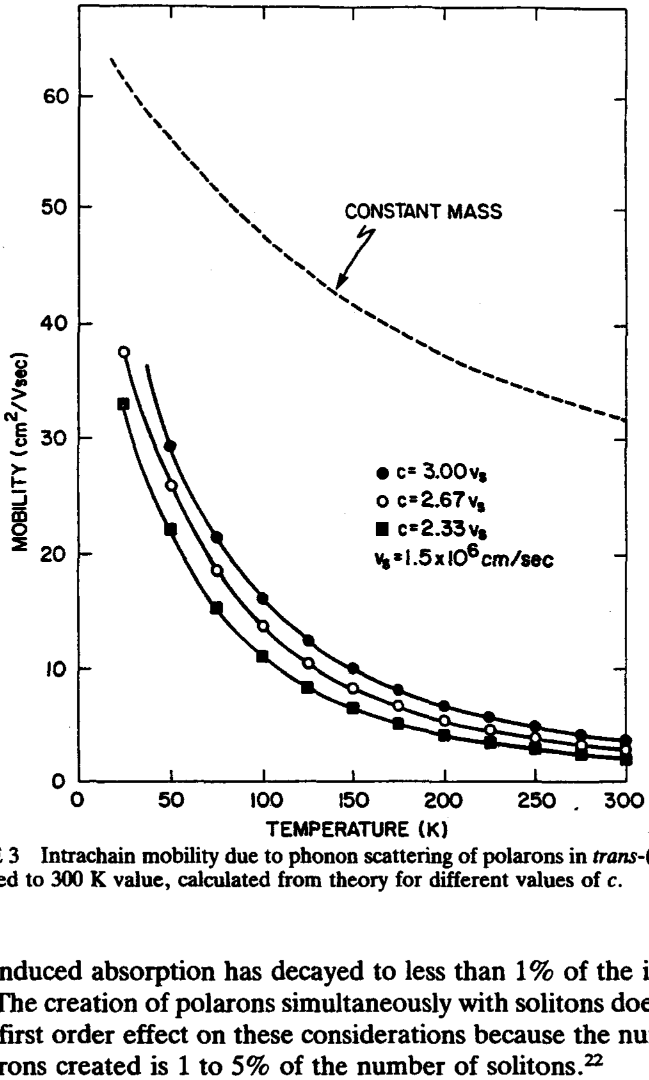

FIGURE 3 Intrachain mobility due to phonon scattering of polarons in *trans*-(CH)$_x$, normalized to 300 K value, calculated from theory for different values of c.

photo-induced absorption has decayed to less than 1% of the initial value. The creation of polarons simultaneously with solitons does not have a first order effect on these considerations because the number of polarons created is 1 to 5% of the number of solitons.[22]

Sinclair *et al.* reported that the 50 ps photoconductivity does not depend much on ambient temperature from 10 to 300 K.[7] A similar finding was made in investigations of photoconductivity nanoseconds after the exciting pulse.[8,9] It was suggested that this finding indicates that the photo-produced carriers are hot.[7,8] Indeed, the carriers are produced with greater than thermal energy. It is straightforward to show, however, with the theory of section 2, that the rate of energy loss is so large that the carriers will thermalize in less than a ps.[23]

In calculating the excess energy of creation it is generally considered that the photon energy in excess of $2\Delta_o$ must be dissipated by the electron and/or hole before the process of soliton or polaron formation begins. (Consistent with this, at photon energies above ~2 eV the 50 ps photoconductivity is independent of photon energy.)[7] The excess energy with which a pair of solitons is formed is then $2\Delta_o - 4\Delta_o/\pi$. With $\Delta_o \simeq 0.7$ eV, this excess energy is $\simeq$0.51 eV. For the solitons that survive long enough to separate, presumably only a fraction, numerical calculations[14] indicate that ~0.37 eV of the excess 0.51 eV has been left behind in a well-localized breather halfway between them. This leaves each soliton with an excess energy of ~0.07 eV minus thermal energy at the ambient temperature. A polaron is left with the excess energy $\Delta - 2\sqrt{2}\Delta/\pi$, which is also 0.07 eV.

The rate of energy loss to acoustic phonons is[23]

$$\frac{dE}{dt} = \frac{\hbar\omega_{em}}{\tau_{em}} - \frac{\hbar\omega_{abs}}{\tau_{abs}} \tag{18}$$

where $\hbar\omega_{em}$ is the average energy of the phonon emitted in mean free time τ_{em}, $\hbar\omega_{abs}$ and τ_{abs} the corresponding quantities for absorption. Calculating these quantities as described in section 2 we obtain the rates of energy loss vs. excess energy shown in Figure 4. The results shown are for 66 K but do not change much with temperature over the range concerned because the loss is dominated by spontaneous emission. According to Figure 4 the time required to lose 0.07 eV is less than 0.1 ps for either soliton or polaron. Even if each soliton had retained half of the 0.51 eV attributed to breather formation this energy would have been lost to acoustic phonon generation in a fraction of a ps. We conclude that with thermalization times less than 1 ps the photoconductivity σ_{ph} observed over a 50 ps interval after the exciting pulse must represent that of solitons and polarons with thermal energy.[23]

In searching for the reason for the weak temperature dependence observed for σ_{ph}, it is important to note that, at least for the lower temperatures investigated, sample temperatures are higher than ambient due to the large energy in the laser pulse and the poor thermal conductivity of polyacetylene at low temperatures.[22] It is not implausible that for a 10 K ambient temperature the sample temperature would be 60 or 70 K. If that were the case, the conclusion from the Sinclair *et al.* measurements[7] would be that σ_{ph} does not depend much on temperature T from ~60 or 70 K to somewhat over 300 K. That conclusion would be in reasonable agreement with our theory. The

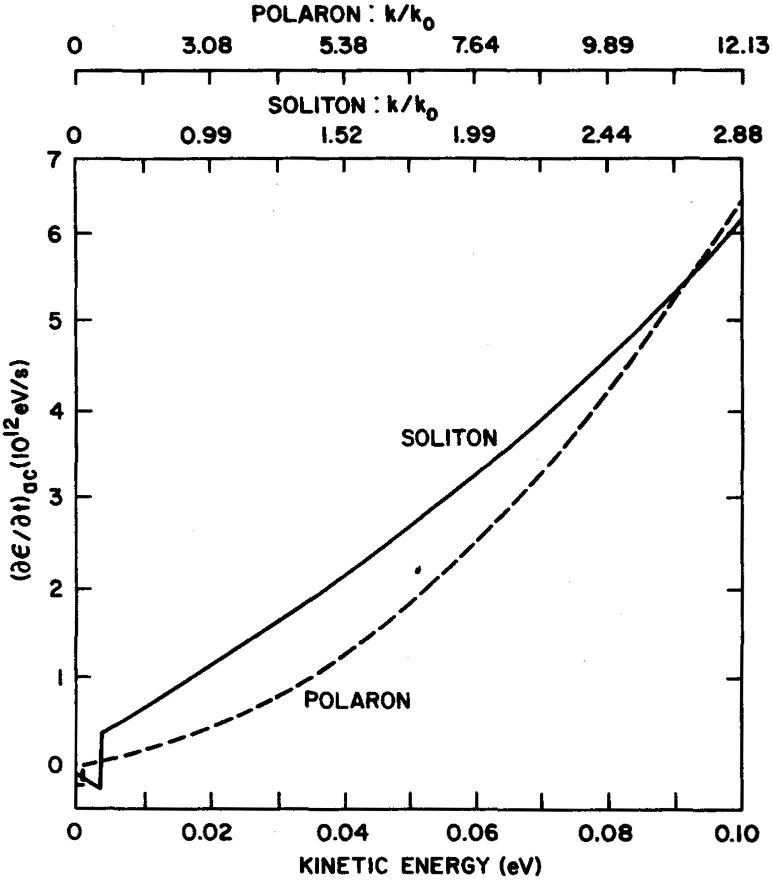

FIGURE 4 Rate of energy loss of solitons and polarons in *trans*-polyacetylene vs. kinetic energy at a lattice temperature of 66 K for maximum velocity $c = 2.67\ v_s = 4 \times 10^6$ cm/s. The scales at the top give the wave vector k in units of $k_o = m^o c/\hbar$, where m^o is the limiting low-velocity mass ($= m_e$ for the polaron, $5.5\ m_e$ for the soliton, m_e being the electron mass).

T-dependence of σ_{ph} arises from (1) the mobility of the carriers and (2) the number of carriers, thus their lifetime. As shown in Figure 7, over the range 70 to 300 K the T-dependence of $\mu_{\parallel}$ for solitons is not large. It is reasonable that this would also be true for polarons drifting parallel to the chains. It would not necessarily be true for interchain hopping of polarons, but in an unoriented sample, as will be seen below, the hopping contribution to σ_{ph} would probably be negligible. So far as lifetime affects on σ_{ph} are concerned, polaron decay, whatever its T-dependence, will not have much effect on $\sigma_{\|}$ because the polarons are converted to charged solitons with about the same $\mu_{\|}$. Soliton decay, as seen in the decay of photoinduced absorption,[5,24] is ~T-independent. Thus the observation of small T-dependence for the 50 ps σ_{ph} would be easily explainable if the actual T of the "10 K" sample were ~60 or 70 K. If the actual T is not that high, it could indicate that polarons dominate the low T σ_{ph}. As suggested earlier, their $\mu_{\|}$ might be higher and less T-dependent than that of solitons because they should be less scattered by conjugation defects.

Photoconductivity measurements on oriented samples have thus far been made only with ns pulses.[8,9] Presumably the ns measurements still represent drifting solitons and perhaps polarons because they show the small dependence on T characteristic of the short-time σ_{ph}.[8,9] It was found[8] that σ_{ph} is higher when the electric vector of the light is perpendicular to the chains than when it is parallel. This had been found earlier for the long-time (μs) σ_{ph}.[25] It is consistent with the preferential creation of polarons when the electric vector is perpendicular to the chains.

Bleier *et al.* investigated also the effect of changing the dc field. For E_{dc} parallel to the chains they found σ_{ph} to be larger by a factor 50 than for E_{dc} perpendicular to the chains.[8] Investigation of the field dependence for E_{dc} parallel to the chains showed it to be mostly sublinear, the exact dependence being a function of light intensity. The sublinear dependence might be due to increased field driving the solitons more rapidly into traps. Also, for fields $>10^4$V/cm carrier heating by the field occurs, causing $\mu_{\|}$ to decrease.[23]

Surprisingly, Bleier *et al.* found no T-dependence of σ_{ph} for E_{dc} perpendicular to the chains.[8] An exponential increase with temperature would be expected because conduction perpendicular to the chains occurs by interchain hopping of polarons.

4. CONCLUSIONS AND FUTURE WORK

Free soliton and polaron diffusion and drift have been observed in *trans*-polyacetylene in a number of different experiments. A theory is available for the transport of these excitations along the chains and has been found to give good agreement with the measured soliton diffusion. Short-time photoconductivity involves both soliton and polaron transport and the interaction between them. The studies made to date have only begun to elucidate the characteristics of the excitations and their interactions in this situation.

It is clear that further photoconductivity measurements on oriented samples, with shorter pulses (down to 1 ps), are needed to learn more about polaron and soliton transport. To separate the contributions of polarons from solitons σ_{ph} should be studied for E_{dc} perpendicular to the chains. If a characteristic optical absorption for the polaron were found, it would be possible to separate soliton and polaron contributions for E_{dc} parallel to the chains by simultaneously monitoring both of their characteristic absorptions. Another possibility for separating soliton and polaron contributions is by study of σ_{ph} in fields greater than 10^4/V/cm. In such fields polaron mobility is predicted[23] to decrease more rapidly with field than soliton mobility because the polaron mass increases more rapidly. It would also be of interest to observe σ_{ph} in samples with very small neutral soliton concentration. Finally, it would increase our knowledge of transport to study σ_{ph} in samples with known densities of conjugation defects to determine their scattering effect on solitons and polarons.

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