Soliton diffusion in polyacetylene. I. Optical phonons

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In this series of work, the soliton diffusion in polyacetylene is studied theoretically within the framework of the model due to Su, Schrieffer, and Heeger. In general, the soliton interacts with both the optical and the acoustic phonons. In this paper we concentrate on the interaction between the soliton and the optical phonon. We find that at room temperature the optical-phonon interaction provides the soliton diffusion constant consistent with that inferred from the magnetic-resonance experiments in pristine polyacetylene. However, at lower temperatures, the optical-phonon mechanism predicts a diffusion constant, which increased similar to $e^{\beta\omega_0}$ as the temperature (β^{-1}) decreases, where ω_0 is the optical-phonon frequency. This indicates that another diffusion mechanism (i.e., the acoustic-phonon scattering) becomes dominant at a lower temperature.

I. INTRODUCTION

The concept of solitons introduced by Su, Schrieffer, and Heeger[1] (SSH) and others[2,3] appears to be central in correlating a variety of physical properties of pristine and lightly doped polyacetylene. One of remarkable observation is that the soliton in pristine polyacetylene is mobile and undergoes one-dimensional diffusive motion.[4,5] This one-dimensional motion is interpreted as the soliton motion along the $(CH)_x$ chain.

The object of this series of work is to study theoretically the soliton diffusion within the SSH model. We shall see in Sec. II that the soliton couples linearly to both optical and acoustic phonons. However, since the coupling constant to the acoustic phonon is smaller than that of the optical phonon by a factor of $a/\xi \sim \frac{1}{7}$, where a is the distance between two adjacent (CH) groups projected along the $(CH)_x$ chain direction and ξ is the spatial extension of the soliton, we shall limit ourselves here to the soliton interaction with the optical phonon. In the second part of this series we shall analyze the interaction between the soliton and the acoustic phonons.

In a preliminary analysis by the author,[6] the coupling to the optical phonon is considered only in the lowest order. However, such a process becomes inefficient in the temperature region $T < \omega_0$, as most of solitons cannot emit optical phonons in this temperature region,[7] where ω_0 is the optical-phonon frequency. In particular, since $\omega_0 \simeq$ 2000 K in polyacetylene, the above process is no longer available at room temperature. Instead, in this temperature region the multiphonon process dominates the soliton diffusion. Looking into the literature on the polaron mobility in ionic crystals, it was discovered that a necessary formalism to deal with the multiphonon process has already been developed and this formalism is called the Osaka-Schultz-Kadanoff theory.[8,9] Following this prescription,[9] it was found that the soliton diffusion constant is given by

$$D = D_0(e^{\beta\omega_0} - 1) \tag{1}$$

for $T \lesssim \frac{1}{2}\omega_0$, where $\beta = T^{-1}$ and D_0 is a constant of the order of 10^{-2} cm^2/sec for the parameters used in the SSH for polyacetylene.

At room temperature, the above diffusion constant is consistent with that deduced from magnetic resonance experiments.[4,5] However, at lower temperatures Eq. (1) gives a far larger diffusion constant than that determined experimentally.[5] In any case at lower temperatures, the acoustic phonons will play a dominant role in the soliton diffusion, which will be discussed in the following paper (hereafter called paper II).

In the above comparison, a recent experimental result by Shiren *et al.*[10] has been ignored. They have done a beautiful spin-echo experiment in pristine polyacetylene and deduced the spin diffusion constant which is by 10^{-2} smaller than that of other groups.[4,5,11] The origin of this discrepancy

is not understood by this author. However, it can be commented that if the value referred by Shiren *et al.* happened to be true, it would cast grave doubts on the soliton model[12] as it would imply the soliton mean free path at room temperature is 10^{-3} Å.

II. MODEL HAMILTONIAN

We shall start with the SSH Hamiltonian[1] of polyacetylene given by

$$H=-\sum_{n,s} t_{n+1,n}(C^{\dagger}_{n+1,s}C_{ns}+\text{H.c.}) + \tfrac{1}{2}K\sum_{n}(y_{n+1}-y_n)^2+\tfrac{1}{2}M\sum_{n}\dot{y}_n^2 \tag{2}$$

and

$$t_{n+1,n}=t_0-\alpha(y_{n+1}-y_n)\,, \tag{3}$$

where y_n is the displacement of the nth (CH) group along the $(CH)_x$ chain direction, and $C^{\dagger}_n$ and C_n are the creation and annihilation operators for π electron at the nth site. First, let us consider the coupling between a soliton and optical phonons. For this purpose it is convenient to introduce the dimerization order parameter[3] $\Delta(x)$ by

$$\Delta(x)=4\alpha\tilde{y}(x)\,, \tag{4}$$

where $\tilde{y}(x)$ is the continuum limit of the staggered displacement field $\tilde{y}_n=(-1)^n y_n$. Then the perfectly dimerized state is given by $\Delta(x)=\pm\Delta$, where 2Δ is the Peierls energy gap. Furthermore, a soliton is described by[3]

$$\Delta_s(x)=\pm\Delta\tanh(x/\xi) \tag{5}$$

with $\xi=v_F/\Delta$.

In the following we shall consider a time-dependent solution:

$$\Delta_s(x,t)=\Delta\tanh[(x-vt)/\xi] \tag{6}$$

which describes a soliton with a uniform velocity v. This moving soliton has energy[1,3]

$$E_s(v)=E_s+\tfrac{1}{2}mv^2\,, \tag{7}$$

where $E_s=(2/\pi)\Delta$ and m is the soliton mass of the order of the electron mass[13] m_e. The moving soliton couples to both the electron and phonon degrees of freedom. However, the former coupling introduces the correction terms of the order of $(v/v_F)^2$, which is completely negligible.[3] Therefore, we shall concentrate on the coupling to phonons. In the case of the optical phonon, this arises from the last term in Eq. (2), which is written in the continuum limit as

$$H_1'=\tfrac{1}{2}g^{-2}\int\dot{\Delta}(x)^2dx\,, \tag{8}$$

where $g=4\alpha(a/M)^{1/2}$ and a dot on $\Delta(x)$ means the time derivative.

Now writing $\Delta(x,t)$ as

$$\Delta(x,t)=\Delta_s(x,t)+\phi(x-vt,t) \tag{9}$$

and substituting it in Eq. (8), we find

$$H_1'=\frac{1}{2g^2}\int dx\left[\frac{v}{\xi}\Delta\,\text{sech}^2\left[\frac{x-vt}{\xi}\right]+\phi_t-v\phi_x\right]^2$$

$$=\tfrac{1}{2}mv^2-g^{-2}v^2\Delta\xi^{-1}\int dx\,\text{sech}^2\left[\frac{x-vt}{\xi}\right]\phi_x$$

$$+\frac{1}{2g^2}\int(\phi_t-v\phi_x)^2dx\,, \tag{10}$$

where subscripts t and x mean the partial derivative. In deriving Eq. (10), we have made use of the orthogonality relation[14]

$$\int dx\,\text{sech}^2\left[\frac{x-vt}{\xi}\right]\phi_t=0\,. \tag{11}$$

From the second term of Eq. (10) the linear coupling term is finally given, which is rewritten as

$$H_1=-ig^{-2}\Delta(2LM\omega_0)^{-1/2}\times\sum_k vv'kf(k)(a_k^{\dagger}-a_{-k}) \tag{12}$$

and

$$f(k)=\xi^{-1}\int dx\,\text{sech}^2\left[\frac{x}{\xi}\right]e^{ikx}$$

$$=\pi\xi k\,\text{csch}\left[\frac{\pi}{2}\xi k\right]\,, \tag{13}$$

where L is the length of the $(CH)_x$ chain, v and v' are the initial and the final velocity of the soliton.

Here $a_k^{\dagger}$ and a_{-k} are the creation and the annihilation operators for the optical phonons and v^2 in the coupling constant is symmetrized as vv'. In general v^2 may be symmetrized as $\frac{1}{2}(v^2+v'^2)$, $\frac{1}{4}(v+v')^2$, etc. However, in the present context, we know that the soliton with $v=0$ does not couple to the optical phonon. Likewise it is so also for $v'=0$. This leads uniquely to the form given in Eq. (12). Also in evaluating the matrix element in Eq. (12), a plane-wave solution for propagating phonons are used instead of a scattering wave solution in the presence of a soliton.[15] It is difficult to

evaluate the error involved here as the exact scattering wave solution is not known in the SSH model. However, if we substitute the one for the ϕ^4 theory[16] (as we know that the ϕ^4 model is quite similar to the SSH model[15]), the coupling constant is reduced by 25% in the limit k tends to zero. Therefore, we may conclude that semiquantatively the present approximation is justified. Furthermore, the k dependence[15] of the optical-phonon frequency $\omega_0(k)$ shall be neglected in the following for simplicity.

The coupling term to the acoustic phonon, on the other hand, comes from the first term in Eq. (1). In the continuum limit, the coupling between the acoustic phonon and the electron is written as

$$H_2 = i\alpha a^2 \sum_s \int dx [\partial_x y(x)] \Psi_s^\dagger \sigma_3 \overleftrightarrow{\partial} \Psi_s \ , \tag{14}$$

where $y(x)$ is the continuum limit of the displacement field y_n, σ_3 is the Pauli spin matrix, and

$$\Psi_s(x) = \begin{bmatrix} u_s(x) \\ v_s(x) \end{bmatrix}$$

is the spinor field consisting of the right-going electron $u(x)$ and the left-going electron $v(x)$.

In the perfectly dimerized state, the expectation value of $i\sum_s \Psi_s^\dagger \sigma_3 \overleftrightarrow{\partial} \Psi_s$ is a constant independent of the position and the above coupling term becomes a complete integral; there is no linear coupling to the acoustic phonon. In the presence of a soliton, on the other hand, we obtain

$$i \sum_s \langle \Psi_s^\dagger \sigma_3 \overleftrightarrow{\partial}_x \Psi_s \rangle = C + \frac{2}{L} \sum_n \frac{1}{\epsilon_n} \partial_x \Delta(x)$$
$$= C + g^{-2} \omega_Q^2 \partial_x \Delta(x) \ , \tag{15}$$

where C is the same constant as in the perfectly dimerized state, $-\epsilon_n$ is the energy of the valence electron, and the summation over n runs up to the Fermi level. A derivation of Eq. (15) will be sketched in the Appendix. Substituting Eq. (15) into Eq. (14) we obtain

$$H_2 = \alpha a^2 g^{-2} \omega_Q^2 \int dx \, \partial_x y \partial_x \Delta(x) \ . \tag{16}$$

This yields

$$H_2 = i g^{-1} C \Delta (2L)^{-1/2}$$
$$\times \sum_k (C \mid k \mid)^{1/2} f(k) (a_k^\dagger - a_{-k}) \ , \tag{17}$$

where C is the acoustic-phonon velocity, $f(k)$ has the same function as defined in Eq. (13), and $a_k^\dagger$ and a_{-k} are now the creation and annihilation operators for the acoustic phonon. In deriving Eq. (17), we made use of the relation[15] $C = \frac{1}{2}\omega_Q a$. The fact that the coupling constant does not vanish in the limit the soliton velocity v tends to zero, implies that a soliton introduces a permanent distortion on the (acoustic) phonon lattice.[17] For a moving soliton with a velocity v, the distorted $y(x)$ field is calculated as

$$y_0(x,t) = -\left[1 - \frac{v^2}{C^2}\right]^{-1} u_0 \tanh\left[\frac{x - vt}{\xi}\right]$$
$$+ C(x - vt) \tag{18}$$

with

$$u_0 = \Delta/(4\alpha)$$

and C is a constant.

As is easily seen, the present coupling constant is of the order of (a/ξ). Therefore, in the continuum limit,[3] the coupling to the acoustic phonon drops out completely from the theory. However, as we shall see in paper II, the acoustic phonon will dominate the soliton diffusion in the low-temperature region.

III. SOLITON RELAXATION

We shall calculate the soliton diffusion constant in two steps: First, we shall determine the transport lifetime of the soliton with momentum $\vec{p}$, and then the diffusion constant is given by $D = \langle \tau v^2 \rangle$, where the angle brackets mean the thermal average. As already mentioned in the temperature region $T < \omega_0$, where the average kinetic energy of the soliton is smaller than the optical-phonon frequency ω_0, the multiphonon process dominates the soliton relaxation. Indeed, we can follow closely an analysis done by Langreth[9] in his calculation of polaron mobility. Introducing the T matrix for the scattering of a soliton of momentum $\vec{p}$ by a phonon of momentum $\vec{k}$ by $T_{\vec{p},\vec{k},\vec{p}',\vec{k}'}$, the transport lifetime $\tau(\vec{p})$ for soliton of momentum $\vec{p}$ is given by[9]

$$\tau^{-1}(p) = \bar{N} \sum_{\vec{p}',\vec{k},\vec{k}'} | T_{\vec{p}',\vec{k}',\vec{p},\vec{k}} |^2$$
$$\times 2\pi \delta(E_p - E_{p'}) \left[1 - \frac{\vec{p}\cdot\vec{p}'}{p^2}\right] , \tag{19}$$

where

$$E_p = E_s + \frac{1}{2m}p^2, \quad \bar{N} = (e^{\beta\omega_0} - 1)^{-1} . \tag{20}$$

Here we have neglected the phonon dispersion in ω_0, which has a minor effect in the final result. Furthermore, the T matrix in Eq. (19) is approximated by[9]

$$T_{\vec{p}\,\vec{k},\vec{p}\,',\vec{k}\,'} = \frac{\delta_{p'+k',p+k} V_{p,p+k} V_{p+k,p'}}{E_p + \omega_0 - E_{p+k} - \sigma} , \tag{21}$$

where

$$V_{p,p'} = ig^{-1}\Delta \left[\frac{p}{m}\right]\left[\frac{p'}{m}\right](2L\omega_0)^{-1/2} \times f(p-p')\,|p-p'| \tag{22}$$

and

$$\sigma \simeq -i\pi \sum |V_{p+k,p+k-q}|^2 \delta(E_p - E_{p+k-q}) . \tag{23}$$

Here Eq. (22) follows from Eq. (12), and p and p' refer to the initial and the final momentum of the soliton. Also, it is known that the approximation Eq. (23) is valid in the weak coupling limit. Therefore, our analysis is also valid in the weak coupling limit. Within this approximation, $|T_{p'k',pk}|^2$ is further approximated by

$$\sum_{kk'} |T_{p'k',pk}|^2 = 2\pi \sum_k \frac{|V_{p,p+k}|^2 |V_{p+k,p'}|^2}{\gamma} \times \delta(E_p + \omega_0 - E_{p+k}) \tag{24}$$

with

$$\gamma = -2\partial m\sigma .$$

Substitution into Eq. (19) gives

$$\tau^{-1}(p) = 2\pi\bar{N} \sum_{p'} \delta(E_p - E_{p'}) \left[1 - \frac{\vec{p}\cdot\vec{p}\,'}{p^2}\right] \sum_{\bar{p}} \frac{|V_{p,\bar{p}}|^2 \delta(E_p - E_{\bar{p}} + \omega_0) |V_{\bar{p},p'}|^2}{\sum_q |V_{\bar{p},q}|^2 \delta(E_{\bar{p}} - E_q - \omega_0)} . \tag{25}$$

Then the summations over q, $\bar{p}$, and p' in Eq. (25) are replaced by integrals and we find

$$\tau^{-1}(p) = 4\bar{N}\left[\frac{\Delta}{g}\right]^2 \left[\frac{p}{m}\right]^2 \bar{p} \left[\frac{p_0^2 |f_1|^2 |f_2|^2}{(p+\bar{p})^2 |f_1|^2 + (p-\bar{p})^2 |f_2|^2}\right] = 2\bar{N}\left[\frac{\Delta}{g}\right]^2 \left[\frac{p}{m}\right]^2 p_0 |f_0|^2 F(p) , \tag{26}$$

where

$$\bar{p} = (p^2 + p_0^2)^{1/2}, \quad p_0 = (2m\omega_0)^{1/2}, \quad f_0 = f(\bar{p}), \quad f_1 = f(\bar{p}+p), \quad f_2 = f(\bar{p}-p) ,$$

and

$$F^{-1} = (2p_0\bar{p})^{-1}[(\bar{p}+p)^2 (f_0/f_1)^2 + (\bar{p}-p)^2 (f_0/f_2)^2] \simeq (\bar{p}/p_0)[1 + 8(p/p_0)^2 + 8(p/p_0)^4]\left\{1 + \left[\frac{\pi}{2}\xi p\right]^2 \left[2 + \operatorname{csch}^2\left[\frac{\pi}{2}\xi\bar{p}\right]\right]\right\} - 4\pi\xi(p^2/p_0)\left[1 + \left[\frac{p}{p_0}\right]^2\right]\left[1 + 2\left[\frac{p}{p_0}\right]^2\right]\tanh\left[\frac{\pi}{2}\xi\bar{p}\right] + O\left[\frac{\pi}{2}\xi p\right]^3 . \tag{27}$$

The transport lifetime $\tau(p)$ of the soliton diverges like p^{-2}, as the momentum of the soliton is reduced.

IV. DIFFUSION CONSTANT

The diffusion constant of the soliton is evaluated as

$$D = \langle \tau(p) v^2 \rangle = \left[\frac{\beta m}{2\pi}\right]^{1/2} \int_{-\infty}^{\infty} dv\, v^2 \tau e^{-(1/2)\beta m v^2} , \tag{28}$$

where $v = p/m$ is the soliton velocity. Here we have assumed that the soliton behaves like a Boltzmann particle. Substituting $\tau(p)$ determined in the preceding section, into Eq. (26), we obtain:

$$D=(2\bar{N})^{-1}\left[\frac{g}{\Delta}\right]^2 p_0^{-1}\,|f_0|^{-2}\langle F^{-1}(p)\rangle$$
$$=\frac{\pi}{4}\left[\frac{\omega_0}{\Delta}\right]\xi\left[\frac{\omega_0}{2m}\right]^{1/2}|f_0|^{-2}\langle F^{-1}\rangle(e^{\beta\omega_0}-1)\,, \tag{29}$$

where

$$\langle F^{-1}\rangle\simeq 1+\tfrac{17}{4}(\beta\omega_0)^{-1}+\left[\frac{\pi}{2}\xi^2\right]\left[\frac{m}{\beta}\right](2+\mathrm{csch}^2\delta)$$
$$-4\pi\xi m(\beta p_0)^{-1}\tanh\delta+O(\beta^{-2}) \tag{30}$$

and

$$\delta=\frac{\pi}{2}\xi p_0\,.$$

Equation (27) can be rewritten as

$$D\simeq D_0(e^{\beta\omega_0}-1)$$

with

$$D_0=\frac{\pi}{4}\left[\frac{\omega_0}{\Delta}\right]\xi\left[\frac{\omega_0}{2m}\right]^{1/2}|f_0|^{-2}\simeq 10^{-2} \tag{31}$$

expressed in units of $\mathrm{cm}^2/\mathrm{sec}$.

Here we made use of accepted values for polyacetylene, $\omega_0=2000$ K, $\Delta=0.7$ eV ($\omega_0/\Delta\simeq 0.25$), $m=3m_e$, and $\xi=7$ Å and m_e is the electron mass. The above diffusion constant is consistent with that deduced from magnetic resonances[4,5] at room temperature. However, at lower temperatures, the predicted diffusion constant increases rapidly as the temperature is decreased, which is in sharp contrast to the experimental observation.[5,11]

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APPENDIX: COUPLING TO THE ACOUSTIC PHONONS

First, let us evaluate

$$\tilde{\rho}(x)=i\sum_s\langle\psi_s^\dagger\sigma_3\overleftrightarrow{\partial}\psi_s\rangle$$

in a uniformly dimerized state. In this case we obtain

$$\tilde{\rho}(x)=\sum_s\int\frac{dp}{2\pi}(2p)(u_p^2-v_p^2)$$
$$=\frac{2}{\pi}\int_{-\Lambda}^{\Lambda}dp\,p\frac{v_Fp}{\epsilon_p}$$
$$=\frac{2}{\pi}v_F^{-2}\left\{\Lambda^2+\frac{\Delta^2}{2}\left[1-\ln\left[\frac{2\Lambda}{\Delta}\right]\right]\right\}=C\,, \tag{A1}$$

where

$$u_p^2=\frac{1}{2}\left[1+\frac{v_Fp}{\epsilon_p}\right],$$
$$v_p^2=\frac{1}{2}\left[1-\frac{v_Fp}{\epsilon_p}\right],$$

and

$$\epsilon_p=[(v_Fp)^2+\Delta^2]^{1/2}\,. \tag{A2}$$

In the presence of a soliton, $\Psi_n(x)$ are given by[3]

$$\Psi_B(x)=\frac{1}{2\sqrt{\xi}}\begin{bmatrix}1\\-i\end{bmatrix}\mathrm{sech}(x/\xi)\,, \tag{A3}$$
$$\Psi_p(x)=\frac{1}{2\sqrt{L}}e^{ipx}\begin{bmatrix}1+\dfrac{v_Fp+i\Delta(x)}{\epsilon_p}\\ i\left[1-\dfrac{v_Fp+i\Delta(x)}{\epsilon_p}\right]\end{bmatrix},$$

where $\Delta(x)=\Delta\tanh(x/\xi)$.

Substituting (A3) into the definition of $\tilde{\rho}(x)$, we obtain

$$\tilde{\rho}(x)=\sum_s\int\frac{dp}{2\pi}\left[p\frac{v_Fp}{\epsilon_p}+\frac{1}{\epsilon_p}\partial_x\Delta(x)\right]$$
$$=C+\frac{1}{\pi}\int_{-\Lambda}^{\Lambda}dp\frac{1}{\epsilon_p}[\partial_x\Delta(x)]$$
$$=C+g^{-2}\omega_Q^2[\partial_x\Delta(x)]\,, \tag{A4}$$

where use is made of a relation

$$1=g^2\omega_Q^{-2}\frac{1}{\pi}\int_{-\Lambda}^{\Lambda}\frac{dp}{\epsilon_p}\,. \tag{A5}$$

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