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Solitons in polyacetylene: Optical absorption in lightly doped polyacetylene

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Optical-absorption spectra of lightly doped *trans*-polyacetylene are studied theoretically within the Su, Schrieffer, and Heeger model and within the dilute-gas approximation for solitons. The present theory describes semiquantitatively the experimental results by Suzuki *et al.*

I. INTRODUCTION

There has been great recent interest in solitons in polyacetylene since the proposal[1,2] that neutral and charged solitons dominate magnetic, electric, and optical properties of undoped and lightly doped polyacetylene. Very recently Suzuki *et al.*[3] have reported a beautiful optical-absorption measurement from lightly doped polyacetylene and have interpreted their experimental data by a theoretical calculation based on the continuum version of the Su, Schrieffer, and Heeger (SSH) model.

Although the calculated absorption coefficient due to the midgap states introduced by solitons appears to describe at least qualitatively the experimental observation, the calculated absorption coefficient associated with the interband transition in the presence of solitons did not appear to describe the experimental observation even qualitatively (the revised version gives a somewhat better description).

The object of the present work is to reexamine theoretically the optical-absorption spectra of *trans*-polyacetylene in the presence of a finite density of solitons. We shall make use of the continuum version of the SSH model formulated by Takayama, Lin-Liu, and Maki (TLM).[4,5]

We shall first show that the interband transition probability in the presence of a single soliton has a spurious infrared divergence. This divergence is eliminated if the effects of other solitons in the same chain are included; the finite concentration of solitons introduces the topological disorder in the chain with the coherence length inversely proportional to the soliton density n_s, which provides a natural infrared cutoff in the theory. This situation is very similar to that discussed by Krumhansl and Schrieffer[6] in the case of the ϕ^4 model.

The resulting optical-absorption spectra appear to describe reasonably well the observed optical-absorption spectra. Indeed we can infer the soliton density in the lightly doped polyacetylene by comparing the theory with the observed optical density. It appears that the soliton densities associated with the optical-absorption spectra[3] are always larger than the dopant concentration by almost a factor of 2, if we assume that each dopant contributes a single soliton to polyacetylene. Furthermore, the soliton densities are not linear in C, the dopant concentration, but have more complicated dependence on C, which may be partly due to the inhomogeneity of doping. In any case, however, the above experimental results demonstrate that the soliton densities increase steadily with doping up to $C \simeq 0.5\%$. In order to construct a theory which describes quantitatively the experimental observation, the inclusion of the lattice fluctuation as well as the randomness in the bond length associated with the bending of the $(CH)_x$ chain seems to be necessary. These questions will be addressed in future publications.

II. FORMULATION

The optical-absorption coefficient is proportional to the electrical conductivity $\sigma(\omega)$. The electrical conductivity is, on the other hand, expressed in terms of the current-current correlation function (Kubo formula) as

$$\sigma(\omega) = \frac{1}{\omega} \operatorname{Im}\{\langle [j,j] \rangle(\omega, 0)\}, \tag{1}$$

where $\langle [j,j] \rangle$ is the retarded product of the current operator $j(z,t)$. Here we are concerned with the conductivity where the current j flows along the CH chain[7] (i.e., $\vec{j}$ parallel to the chain direction).

In order to illustrate the method, let us calculate $\langle [j,j] \rangle$ in the soliton-free case. We assume that the system is described by the Hamiltonian[4,5,8]

$$H_c = \frac{\omega_Q^2}{2g^2} \int dx\, \Delta^2(x) + \sum_s \int dx\, \psi_s^\dagger(x) [-iv_F \sigma_3 \partial_x + \sigma_1 \Delta(x)] \psi_s(x), \tag{2}$$

where

$$g = 4\alpha (a/M)^{1/2}, \quad \omega_Q^2 = 4K/M, \quad \Delta(x) = g(a/M)^{-1/2} \tilde{y}(x), \tag{3}$$

and $\tilde{y}_n = (-1)^n y_n$, the lattice displacement. Equation (2) is the continuum version of the SSH model, where $\psi(x) \equiv \binom{u(x)}{v(x)}$ is the spinor representation of the electron field.[9] The soliton-free dimerized state corresponds to $\Delta(x) \equiv \Delta = 4We^{-1/2\lambda}$, where $4W$ is the total bandwidth and $\lambda = g^2/(\pi v_F \omega_Q^2)$ is the effective coupling constant.[10] The electron Green's function is given by

$$G(\omega_n, k) = -(i\omega_n + \sigma_3 v_F k + \sigma_1 \Delta)^{-1} = -(-i\omega_n + \sigma_3 v_F k + \sigma_1 \Delta)(\omega_n^2 + E_k^2)^{-1}, \quad (4)$$

where

$$E_k = [(v_F k)^2 + \Delta^2]^{1/2} \quad (5)$$

and $\omega_n = 2\pi T(n + \frac{1}{2})$ is the Matsubara frequency for the fermion.

Making use of the fact that the current operator is given by

$$j(x) = ev_F \sum_s \psi_s^\dagger(x)\sigma_3\psi_s(x), \quad (6)$$

the retarded product of the current operator is given by

$$\langle [j,j] \rangle_0(i\omega_\nu) = 2T(ev_F)^2 \sum_n \int \frac{dk}{2\pi} \mathrm{Tr}[\sigma_3 G(\omega_n, k) \times \sigma_3 G(\omega_{n-\nu}, k)] \quad (7)$$

by analytical continuation. At $T = 0$ K then, Eq. (7) is easily evaluated as

$$\langle [j,j] \rangle(\omega) = \frac{2e^2 v_F}{\pi}\left(1 - v_F \tfrac{1}{2} \int_{-\infty}^{\infty} dk \frac{\Delta^2}{E_k[E_k^2 - (\frac{1}{2}\omega)^2]}\right) = \frac{2e^2 v_F}{\pi}\left(1 - \frac{1}{z(z^2-1)^{1/2}} \sin^{-1} z\right) \quad \text{for } |z| < 1 \quad (8)$$

$$= \frac{2e^2 v_F}{\pi}\left[1 - \frac{1}{z(z^2-1)^{1/2}}\left(\frac{\pi}{2} i - \cosh^{-1} z\right)\right] \quad \text{for } |z| > 1, \quad (9)$$

where $z = \omega/2\Delta$. The frequency-dependent conductivity from a single chain of *trans*-polyacetylene is given by

$$\sigma(\omega) = \frac{e^2 v_F}{\Delta} z^{-2}(z^2-1)^{-1/2}\Theta(|z| - 1), \quad (10)$$

which was obtained previously by SSH.[1]

III. CONDUCTIVITY IN THE PRESENCE OF SOLITONS

In order to study the effects of solitons on $\sigma(\omega)$, we shall first consider a polyacetylene with a single soliton at $x = x_0$. In this case the Green's function is given by

$$G(\omega_n, k) = \frac{1}{i\omega_n - E_k}\begin{pmatrix} u_k^*(x)u_k(y) & u_k^*(x)v_k(y) \\ v_k^*(x)u_k(y) & v_k^*(x)v_k(y) \end{pmatrix} + \frac{1}{i\omega_n + E_k}\begin{pmatrix} v_k^*(x)v_k(y) & -v_k^*(x)u_k(y) \\ -u_k^*(x)v_k(y) & u_k^*(x)u_k(y) \end{pmatrix} - i\delta_{k0}\frac{2}{\omega_n} u_B(x)u_B(y)\begin{pmatrix} 1 & -i \\ i & 1 \end{pmatrix}, \quad (11)$$

where

$$u_k(x) = \frac{1}{2}\left(1 + \frac{v_F k + i\Delta(x)}{E_k}\right), \quad v_k(x) = \frac{i}{2}\left(1 - \frac{v_F k + i\Delta(x)}{E_k}\right), \quad (12)$$

$$u_B(x) = iv_B(x) = \frac{1}{\sqrt{4\xi}} \mathrm{sech}\left(\frac{x - x_0}{\xi}\right),$$

and

$$\Delta(x) = \Delta \tanh\left(\frac{x - x_0}{\xi}\right). \quad (13)$$

Here we have made use of the electron wave functions given in TLM.[4]

The retarded product of the current operators for a single polyacetylene chain is calculated as

$$\langle [j,j] \rangle(i\omega_\nu) = 2T(ev_F)^2 \int_0^L \frac{dx_0}{L} \sum_n \int \frac{dk}{2\pi} \int \frac{dk'}{2\pi} e^{i(k-k')(x-y)} \mathrm{Tr}[\sigma_3 G(\omega_n, k)\sigma_3 G(\omega_{n-\nu}, k')], \quad (14)$$

where now $G(\omega_n, k)$ is given by Eq. (11) and we have averaged over the soliton position x_0. Here L is the length of the $(CH)_x$ chain. Then Eq. (14) is rewritten as

$$\langle [j,j] \rangle(\omega) = \frac{2e^2 v_F}{\pi}[1 - I^{(1)}(\omega) - I^{(2)}(\omega)], \quad (15)$$

where

$$I^{(1)}(\omega)=v_F\int_{-\infty}^{\infty}dk\,|M_k^{(1)}|^2\,\frac{2E_k}{E_k^2-\omega^2}, \tag{16}$$

$$I^{(2)}(\omega)=\frac{v_F}{2\pi}\int_{-\infty}^{\infty}dk\int_{-\infty}^{\infty}dk'\,|M_{k-k'}^{(2)}|^2\,\frac{2(E_k+E_{k'})}{(E_k+E_{k'})^2-\omega^2}\ , \tag{17}$$

$$M_k^{(1)}=\frac{1}{(4\xi L)^{1/2}}\int_{-\infty}^{\infty}dx\,\mathrm{sech}\left(\frac{x}{\xi}\right)e^{ikx}=\frac{\pi}{2}\left(\frac{\xi}{L}\right)^{1/2}i\,\mathrm{sech}\left(\frac{\pi}{2}k\xi\right), \tag{18}$$

and

$$M_{k-k'}^{(2)}=\frac{1}{\sqrt{L}}\int_{-\infty}^{\infty}dx[u_k(x)v_{k'}^*(x)+v_k(x)u_{k'}^*(x)]\,e^{i(k-k')x}=\frac{i}{2\sqrt{L}}\int_{-\infty}^{\infty}dx\left[\frac{v_Fk}{E_k}-\frac{v_Fk'}{E_{k'}}+i\Delta(x)\left(\frac{1}{E_k}+\frac{1}{E_{k'}}\right)\right]e^{i(k-k')x}$$

$$=\frac{i\pi}{\sqrt{L}}\left[\delta(k-k')\left(\frac{v_Fk}{E_k}-\frac{v_Fk'}{E_{k'}}\right)-\frac{\Delta}{2}\left(\frac{1}{E_k}+\frac{1}{E_{k'}}\right)\xi\,\mathrm{csch}\left(\frac{\pi}{2}(k-k')\xi\right)\right]. \tag{19}$$

Here $I^{(1)}(\omega)$ is associated with the midgap transition, while $I^{(2)}(\omega)$ describes the interband transition.

Taking the imaginary part of Eq. (15), the conductivity in the presence of a single soliton is given by

$$\sigma(\omega)=\sigma^{(1)}(\omega)+\sigma^{(2)}(\omega), \tag{20}$$

where

$$\sigma^{(1)}=(e^2v_F^2)\,\frac{\xi}{\omega L}\,\pi^2\int_{-\infty}^{\infty}dk\,\mathrm{sech}\left(\frac{\pi}{2}k\xi\right)\delta(E_k-\omega)=\frac{e^2v_F}{2\Delta}\left(\frac{\xi}{L}\right)\pi^2(z^2-\tfrac{1}{4})^{-1/2}\,\mathrm{sech}^2[\pi(z^2-\tfrac{1}{4})^{1/2}]\,\Theta(|z|-\tfrac{1}{2})\ , \tag{21}$$

$$\sigma^{(2)}(\omega)=\frac{e^2v_F^2}{4\omega L}\,\pi^2\int_{-\infty}^{\infty}dk\int_{-\infty}^{\infty}dk'\left[\delta(k-k')\left(\frac{v_Fk}{E_k}-\frac{v_Fk'}{E_{k'}}\right)-\frac{\Delta}{2}\left(\frac{1}{E_k}+\frac{1}{E_{k'}}\right)\xi\,\mathrm{csch}\left(\frac{\pi}{2}(k-k')\xi\right)\right]^2\delta(E_k+E_{k'}-\omega), \tag{22}$$

where Eq. (21), which describes the midgap transition, has been correctly given by Suzuki *et al.*[3] On the other hand, Eq. (22), which describes the interband transition, diverges because of the pole at $k=k'$. This divergence is eliminated by introducing the effect of other solitions, which gives rise to a finite coherence length $\xi_c \equiv a(2n_s)^{-1}$, where a is the distance between two adjacent (CH) groups measured along the $(CH)_x$-chain direction and n_s is the soliton density. A detailed discussion of the topological disorder due the a finite soliton density will be described in Appendix A. With this in mind replace the pole in Eq. (22) by

$$(k-k')^{-2}\to[(k-k')^2+(2n_sa^{-1})^2]^{-1}\ ,$$

etc. Then Eq. (22) is rewritten as

$$\sigma^{(2)}(\omega)=\frac{e^2v_F^2}{\omega L}\int_{-\infty}^{\infty}dk\int_{-\infty}^{\infty}dk'\frac{1}{[(k-k')^2+(2n_sa^{-1})^2]}\left[\pi\left(\frac{v_Fk}{E_k}-\frac{v_Fk'}{E_{k'}}\right)\frac{2n_sa^{-1}}{(k-k')}-\Delta\left(\frac{1}{E_k}+\frac{1}{E_{k'}}\right)\right]^2\delta(E_k+E_{k'}-\omega)$$

$$=\frac{e^2v_F}{\pi\Delta}\,\frac{4\xi}{L}\,z\int_0^{(z^2-1)^{1/2}}\frac{dy}{K^2+y^2}\,\frac{1}{[(z^2-y^2)^2+y^2]}\left(\frac{z^2-y^2}{z^2-y^2-1}\right)^{1/2}\left(1-\frac{K}{z^2-y^2}\right)^2\ , \tag{23}$$

where

$$z=\omega/2\Delta,\quad K=n_s\xi a^{-1}\ . \tag{24}$$

Finally, in the presence of a finite density of soliton, we have

$$\sigma(\omega)=\frac{e^2v_F}{\Delta}\,K\left[\Theta(|z|-\tfrac{1}{2})\,\frac{\pi^2}{2}\,(z^2-\tfrac{1}{4})^{-1/2}\,\mathrm{sech}^2[\pi(z^2-\tfrac{1}{4})^{1/2}]\right.$$

$$\left.+\,\Theta(|z|-1)\,\frac{2z}{\pi}\int_0^{(z^2-1)^{1/2}}dy\,\frac{1}{K^2+y^2}\,\frac{1}{(z^2-y^2)^2+y^2}\left(\frac{z^2-y^2}{z^2-y^2-1}\right)^{1/2}\left(1-\frac{K}{z^2-y^2}\right)^2\right], \tag{25}$$

which is obtained by multiplying $n_s La^{-1}$ on Eq. (20).

The above expression is numerically evaluated for several n_s's and shown in Fig. 1. We have taken $\xi/a = 7$ in the numerical calculation. As is easily seen, the present results describe semiquantitatively the observed optical spectra of lightly doped polyacetylene for the whole frequency range, if we ignore the square-root singularities at the thresholds of the midgap transition as well as the interband transition. These singularities may be washed away, if the effects of the thermal phonons are included in the theoretical calculation.

However, comparing the relative weight of the absorption due to the midgap transition to the total absorption, it appears that the soliton densities n_s are consistently larger than the dopant concentration C, if each dopant introduces one soliton to polyacetylene. For example, we obtain $n_s = 0.08$, 0.16, 0.42, and 0.91% for $C = 0$, 0.01, 0.1, and 0.5%, respectively, by analyzing the optical-absorption spectra reported by Suzuki *et al.*[3] In particular, $n_s = 0.08$ for undoped polyacetylene appears to be consistent with magnetic susceptibility measurements in pristine *trans*-polyacetylene.[11-13] Furthermore, if we take the present analysis at face value, a single dopant appears to create more than one soliton for example, one and a half solitons on the average. This may be plausible as the soliton cannot be created singly in a single $(CH)_x$ chain due to the topological constraint.

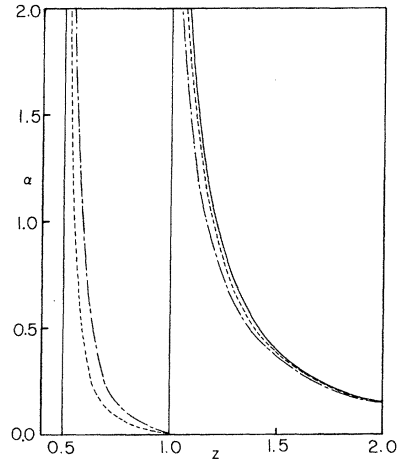

FIG. 1. The optical-absorption spectra are calculated as function of reduced optical frequency $z = \omega/(2\Delta)$ for the soliton density $n_s = 0$ (solid curve), $n_s = 1\%$ (broken curves) and $n_s = 2\%$ (chained curve). Note that the midgap absorption increases linearly with n_s.

In the limit of small n_s, Eq. (25) may be approximated as

$$\sigma(\omega) \cong \frac{e^2 v_F}{\Delta}\left(K\,\frac{\pi^2}{2}\,(z^2-\tfrac{1}{4})^{-1/2}\,\mathrm{sech}^2[\pi(z^2-\tfrac{1}{4})^{1/2}]\,\Theta(|z|-\tfrac{1}{2}) + \Theta(|z|-1)z^{-2}(z^2-1)^{-1/2}(1-2Kz^{-2}) + O(K^2)\right), \quad (26)$$

where $z = \omega/(2\Delta)$ and $K = n_s \xi a^{-1}$.

This is a reasonable approximation for $K \lesssim 10^{-2}$ (or $n_s \lesssim 0.1\%$) and z not far from the interband threshold ($z - 1 \lesssim 1$). The interband term in the above expression is similar to the one in the revised version of Suzuki *et al.*[3] but differs in details. Furthermore, the present expression appears not to satisfy the optical sum rule. This is easily seen from

$$\frac{\pi^2}{2}\int_{1/z}^{\infty} dz\,(z^2-\tfrac{1}{4})^{-1/2}\,\mathrm{sech}^2[\pi(z^2-\tfrac{1}{4})^{1/2}] = 2.82\ldots$$

and

$$2\int_1^{\infty} dz\, z^{-4}(z^2-1)^{-1/2} = \tfrac{4}{3} = 1.3333\ldots.$$

However, we do not yet understand why the sum rule is not obeyed.

So far we have completely neglected the Coulomb interaction between electrons as in the original SSH model.[1] If the Coulomb interaction were included, this would produce bound states of electron and hole (i.e., excitons) which would introduce an additional exciton structure[14] below $\omega = 2\Delta$. Therefore the optical-absorption experiment will be extemely useful to assess the importance of the Coulomb interaction in polyacetylene.

IV. CONCLUDING REMARKS

Making use of the Green's-function technique, we have calculated the electric conductivity of trans-polyacetylene in the presence of solitons. The present theory appears to account semiquantitavely for the observed optical-absorption spectra by Suzuki *et al.*[3] on *trans*-polyacetylene doped with AsF_5, if the soliton density is assumed slightly larger than that inferred from the dopant concentration. The larger soliton density may arise from the intrinsic solitons, inhomogeneity of doping, and/or the doping mechanism of which detail is not known.

The most significant discrepancies between the present theory and the observed optical spectra

are rounding of the observed spectra near the thresholds of both the midgap transition and the interband transition. We believe that inclusion of the lattice fluctuation (thermal as well as quantum mechanical) will produce sufficient rounding in the absorption spectra near the thresholds. This will be considered in a future publication.

After completing this work we received a report by Gammel and Krumhansl on the optical absorption in polyacetylene with similar results. However, their treatment as well as their expression of the interband absorption are somewhat different from ours.

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APPENDIX: THE TOPOLOGICAL DISORDER IN POLYACETYLENE DUE TO SOLITONS

We shall first consider the correlation of the displacement field $\tilde{y}(x)$ in the presence of a finite soliton density n_s. This can be done as follows. We shall first consider $\tilde{y}(x)$ given by[15]

$$\tilde{y}(x) = y_0 \prod_i \tanh\left(\frac{x - x_i}{\xi}\right), \tag{A1}$$

where x_i are the positions of solitons, then we obtain

$$\langle \tilde{y}(x+y)\,\tilde{y}(x)\rangle = y_0^2 \left\langle \prod_i \tanh\left(\frac{x+y-x_i}{\xi}\right) \times \tanh\left(\frac{x-x_i}{\xi}\right)\right\rangle . \tag{A2}$$

Averaging over x_i's we obtain

$$\langle \tilde{y}(x+y)\,\tilde{y}(x)\rangle = y_0^2\left(1 - \frac{2\xi}{L}\,\frac{y}{\xi}\coth\frac{y}{\xi}\right)^{n_s L/a} \simeq y_0^2 \exp(-2n_s a^{-1}|y|), \tag{A3}$$

where we made use of the relation

$$\frac{1}{L}\int_{-L/2}^{L/2} dx \tanh\left(\frac{x+y}{\xi}\right)\tanh\frac{x}{\xi} = 1 - \frac{2y}{L}\coth\frac{y}{\xi} \tag{A4}$$

and L is the length of the system.

Equation (A3) indicates that the dimerization coherence length is given by $\xi_c = a(2n_s)^{-1}$, which is identical to the case of the ϕ^4 model discussed by Krumhansl and Schrieffer.[6]

If these solitons are moving like the ideal Boltzmann gas, the space-time correlation of the dimerization may be given by[16]

$$\langle \tilde{y}(x,t)\,\tilde{y}(0,0)\rangle = y_0^2 \exp[-2n_s a^{-1}(y^2 + v_0^2 t^2)^{1/2}] \tag{A5}$$

and $v_0 = [(2/\pi)(T/m_s)]^{1/2}$ is the thermal velocity of soliton with mass $m_s \simeq 6m_e$.

We shall now look into a particular term in the calculation of the optical-absorption spectra. We shall consider the integral

$$I(q) = \frac{1}{L}\int_{-L/2}^{L/2} dx \int dy e^{iqy}\Delta(x+y)\Delta(x). \tag{A6}$$

In the case of a single soliton (A6) is evaluated as follows:

$$\begin{aligned} I_1(q) &= \frac{1}{L}\int_{-L/2}^{L/2} dx \int dy\, e^{iqy}\Delta^2 \tanh\frac{x+y}{\xi}\tanh\frac{x}{\xi} \\ &= \Delta^2 \int dy e^{iqy}\left(1 - \frac{2y}{L}\coth\frac{y}{\xi}\right) \\ &= \Delta^2\left[2\pi\delta(q) + \frac{\pi^2}{L}\,\xi\,\mathrm{csch}^2\left(\frac{\pi}{2}q\xi\right)\right] \\ &\simeq \Delta^2\left(2\pi\delta(q) + \frac{4\xi^{-1}}{L}q^{-2}\right). \end{aligned} \tag{A7}$$

The second term has the q^{-2} singularity. In the presence of a finite density of solitons we obtain

$$\begin{aligned} I(q) &= \frac{\Delta^2}{L}\int_{-L/2}^{L/2} dx \int dy\, e^{iqy}\langle \tilde{y}(x+y)\,\tilde{y}(x)\rangle \\ &= \Delta^2 \int dy \exp(iqy - 2n_s a^{-1}|y|) \\ &= \Delta^2 \frac{4n_s a}{q^2 + (2n_s a^{-1})^2}. \end{aligned} \tag{A8}$$

In this way the q^{-2} singularity in the single-soliton term is eliminated when a finite concentration of solitons is considered.

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