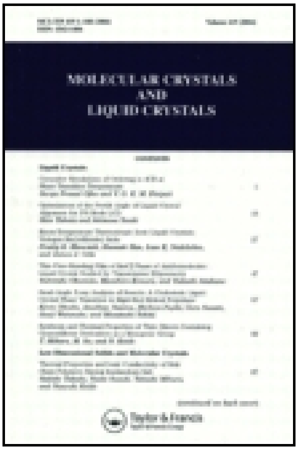

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Infrared Absorption of Doped Polyacetylene

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INFRARED ABSORPTION OF DOPED POLYACETYLENE*

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The frequency dependent conductivity of added charge (e.g. solitons) to a dimerized Peieres condensate with N coupled phonons is studied. The electron-phonon system, with electron-electron interactions and pinning effects, yields in the adiabatic and continuum limits an infrared absorption whose structure is independent of the charge configuration. The kinetic mass of the charge affects only the pinning parameter and the over-all magnitude of the absorption. This mass is estimated from experimental data and compared with theoretical soliton and polaron masses.

The doping process of polyacetylene and the nature of the charge transfer are of considerable interest.[1] In particular Fincher et al. have shown that lightly doped (<0.1%) polyacetylene $(CH)_x$ with a variety of acceptors or donors leads to the appearance of new infra-red (IR) active modes at 900 cm^{-1} (width of ∿400 cm^{-1}) and at 1370 cm^{-1} (width of ∿50 cm^{-1}). This IR activity is independent of the dopant type and is therefore evidence that charge has been transferred to the polyacetylene chain and its coupling with the

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polyacetylene vibrational normal modes causes the IR activity. Of additional interest is the IR data of $(CD)_x$ [3-5] where three lines appear with Na doping.[5]

Undoped polyacetylene is a semiconductor with a gap of $2\Delta_o \approx 1.4$ eV.[6] This gap is maintained upon doping, so that the new IR modes are within the gap, at frequencies $\omega \ll \Delta_o$.

This behaviour is considered as an evidence [4,7] for the soliton configuration in polyacetylene.[8-11] Here I show that this unusual behaviour is a universal result of the translation degree of freedom of the added charge, independent of its configuration. The center of mass coordinate is considered as a time dependent field $\phi(t)$; it is a linear combination of the lattice normal modes which couple to the electrons. By solving the coupled set of equations of motion the IR frequencies ω_n^ϕ $(n=1,2,\ldots,N)$ are obtained as function of the bare phonon frequencies ω_n^o and the dimensionless electron-phonon coupling constants λ_n.

Two assumptions are made in the derivation:

a) All phonon frequencies are small compared with internal electronic transition frequencies. The latter are of order Δ_o and the conditions $(\omega_n^\phi/\Delta_o)^2 \ll 1$ are valid for polyacetylene. In the presence of a pinning potential, the electronic transition frequencies in this potential are also assumed to be large compared with the phonon frequencies.

b) Lattice discreteness is neglected, i.e. the excess charge is delocalized over a distance much longer than the lattice constant a. This assumption is consistent with the metallic behaviour above 1% doping[6] and with the results below.

The infrared conductivity of N localized modes is derived. In principle all modes in the Brillouin zone are infrared active since translation invariance is broken. However, the contribution of extended modes both at the zone center and further than ξ^{-1} from the zone center are both vanishing. Thus for $\xi \gg a$ this contribution is very small, as indeed shown by the numerical lattice calculation.[7]

The approach to intra-gap IR activity, presented below, generalizes two known examples. The first one is the motion of an incommensurate charge density wave[12,13]. The ion displacement at the m-th site is $u(m)=u_o\cos(q_o ma+\phi)$ where u_o is the displacement amplitude, $q_o a/\pi$ irrational and the phase variable ϕ is the center of mass coordinate mentioned above. The second example is the IR activity of charged solitons in polyacetylene.[7]

In a dimerized Peierls condensate, such as polyacetylene, $q_o=\pi/a$ so that $u(m)=(-)^m u_o$; thus ϕ is not coupled to the ion displacement and there are no IR modes (except at high frequencies[14]). The addition of charge, e.g. by doping, restores the IR activity associated with the translation mode, as shown next.

The displacement pattern along the chain axis has the form $u_n(m)=(-)^m\Delta_n(ma)/4\alpha_n$, where $n=1,2,...,N$ are the normal modes and $_n$ their couplings to the electrons as defined in Ref. (8). The neglect of the lattice discreteness leads to a continuum model for $\Delta_n(x)$.[9-11] The Δ_n dependent part of the Hamiltonian is

$$H\left\{\Delta_n\right\} = \int dx \left\{ \sum_{n=1}^{N} (2\pi v_F\lambda_n)^{-1}[\Delta_n^2(x)+(\dot{\Delta}_n(x)/\omega_n^o)^2] + \Delta(x)C(x)\right\} \tag{1}$$

where v_F is the Fermi velocity, $\lambda_n = 4\alpha_n^2 a/(\pi v_F K_n)$ with K_n the spring constants for distorting the undimerized system[8], $\dot{\Delta}_n$ the canonical momenta, $\Delta(x) = \sum_n \Delta_n(x)$ and $C(x)$ involves electron operators describing backscattering of electrons across the Fermi sea. The total Hamiltonian involves also the electron kinetic energy and direct electron-electron interactions. Here only the phonon equations of motion are relevant, and Eq. (1) yields

$$\Delta_n(x,t)+\ddot{\Delta}_n(x,t)/\omega_n^{o2} = -\pi v_F\lambda_n C(x,t) \quad . \tag{2}$$

The ground state of the dimerized system is uniform $\Delta(x)=\Delta_o$. The addition of charge leads to an x dependent solution

$$\Delta_n(x) = -\pi v_F\lambda_n C(x) = \Delta(x)\cdot\lambda_n/\lambda \tag{3}$$

where $\lambda \equiv \sum_n \lambda_n$. Consider now a solution of the form $\Delta(x-\phi(t))$, i.e. the static solution with time dependent center of mass. Each normal mode has its own center of mass variable $\phi_n(t)$ satisfying $\Delta(x-\phi) = \sum_n \Delta_n(x-\phi_n)$. Expanding to first order in ϕ_n yields $\phi(t) = \sum_n \phi_n(t)\lambda_n/\lambda$.

The main ingredient in the derivation is that the electronic part, $C(x,t)$, follows adiabatically the ion displacement, which is justified for $(\omega/\Delta_o)^2 \ll 1$. Thus $\Delta(x-\phi) = -\pi v_F\lambda C(x,t)$ and to first order in $\phi(t)$ it yields $\Delta'(x)\phi = \pi v_F\lambda\delta C$. Expansion of Eq. (2) to first order in ϕ_n yields then

$$\phi_n(t)+\ddot{\phi}_n(t)/\omega_n^{o2}-\phi(t) = 0 . \tag{4}$$

These equations correspond to the Lagrangian

$$L\{\phi_n\} = \tfrac{1}{2} M_c N_c \Omega_o^2 \{\sum_n [-\phi_n^2+(\dot{\phi}_n/\omega_n^o)^2]\lambda_n/\lambda+\phi^2\} \tag{5}$$

where N_c is the charge involved (in units of e),

$$\Omega_o^{-2} = \sum_n \frac{\lambda_n}{\lambda} (\omega_n^o)^{-2} , \tag{6}$$

and M_c the kinetic mass per unit charge. To see this consider a uniform motion with velocity v, i.e. $\phi_n=\phi$, $\dot{\phi}_n=v$ so that $L = \tfrac{1}{2} M_c N_c v^2$.

The mass M_c can be identified from the kinetic term in Eq. (1) where $\dot{\Delta}_n(x-\phi) = \dot{\phi}\Delta_n'(x)$ to first order in ϕ; hence

$$M_c = \int \Delta'^2(x)\,dx/(\pi\lambda v_F \Omega_o^2 N_c) . \tag{7}$$

For the single soliton solution $\Delta(x) = \Delta_o \tanh(x\Delta_o/v_F)$ Eq. (7) yields the known result[8,10] $M_s = 4\Delta_o^3/(3\pi\lambda v_F^2\Omega_o^2)$. For the polaron solution[15]

$$\Delta(x) = \Delta_o-\Delta_o[1+\cosh(\sqrt{2}\ x\ \Delta_o/v_F)/\sqrt{2}]^{-1} . \tag{8}$$

Eq. (7) yields for the ratio of polaron to soliton masses

$$M_p/M_s = 2\sqrt{2} - 3\ln(1+\sqrt{2}) \simeq 0.18 . \tag{9}$$

In the incommensurate limit (i.e. high density of the soliton lattice[16]) $\Delta(x) = \Delta_o \exp(i2k_F x)+H.C.$ where k_F is the Fermi wavevector. Eq. (7) now yields the mass $M_c=M_F$ where $M_F/m = 4\Delta_o^2/\lambda\Omega_o^2$, and $m = k_F/v_F$. M_F is just the Fröhlich mass of an incommensurate charge density wave[17,18] if $\Omega_o^2 \ll \Delta_o^2$.

The motion of the center of mass implies an electric current $j(t) = e\rho\partial\phi/\partial t$ where $\rho=N_c/L$, and L is the length of the system. The charge is assumed to move rigidly with the center of mass, which is again justified for low frequencies $(\omega/\Delta_o)^2 \ll 1$. As an example, a linear response analysis for the soliton solution[9-11] yields

$$j(\omega) = ie\rho\omega\phi(\omega)[1+(\pi^2/12)(\omega/\Delta_o)^2+O(\omega/\Delta_o)^4].$$

The Lagrangian (5) describes a frictionless motion of the charge. The dopant ions however provide a pinning potential for the charge on the polyacetylene chain. To second order in ϕ this defines a pinning parameter α in the effective Lagrangian

$$L_{eff} = \tfrac{1}{2} \cdot M_c N_c \Omega_o^2 \left\{ \sum_n [-\phi_n^2 + (\dot{\phi}_n/\omega_n^o)^2] \lambda_n/\lambda + (1-\alpha)\phi^2 \right\}$$

$$- e^2 N_c A(t) \dot{\phi}(t) \ . \tag{10}$$

and an external electromagnetic potential $A(t)$ is also included. The equations of motion for $\phi_n(\omega)$ are now

$$(1-\omega^2/\omega_n^{o2})\phi_n(\omega) - (1-\alpha)\phi(\omega) = -e^2 E(\omega)/(M_c \Omega_o^2) \tag{11}$$

where $E(\omega)$ is the electric field. Eq. (11) is easily solved for the field $\phi(\omega)$ and Eq. (8) yields the conductivity

$$\sigma(\omega) = i\omega \frac{e^2\rho}{M_c \Omega_o^2} \frac{D_o(\omega)}{1+(1-\alpha)D_o(\omega)} \tag{12}$$

where

$$D_o(\omega) = \sum_n \frac{\lambda_n}{\lambda} \frac{\omega_n^{o2}}{\omega^2 - \omega_n^{o2}} \ . \tag{13}$$

The poles of Eq. (12) yield the IR frequencies ω_n^ϕ. For $\alpha=0$ there is a pole at $\omega=\omega_1^\phi=0$ which corresponds to the Fröhlich type superconductivity[16] $\mathrm{Re}\sigma(\omega) = \delta(\omega)\pi\rho/M_c$ for $\omega<\omega_2$ (ω_2^ϕ is the next pole of Eq. (12)). For $\alpha\neq0$ translation invariance is lost and all modes have a translation mode component with $\omega_n^\phi\neq0$; ω_1^ϕ may be called the "pinned mode", but it must be treated together with all the other modes. The function $D_o(\omega)$ is plotted in Fig. 1 for a 3 phonon system. Its intersections with the value $-1/(1-\alpha)$ (line a in Fig. 1) determine the frequencies ω_n^ϕ. For $\alpha<1$, $\omega_1^\phi<\omega_1^o$ and there is an additional solution in each interval $(\omega_n^o,\omega_{n+1}^o)$. For $\alpha>1$ there is a solution at $\omega>\omega_N^o$ instead of the solution at $\omega<\omega_1^o$. There are always N solutions, i.e. the number of IR modes, including the pinned mode, equals the number of coupled bare modes.

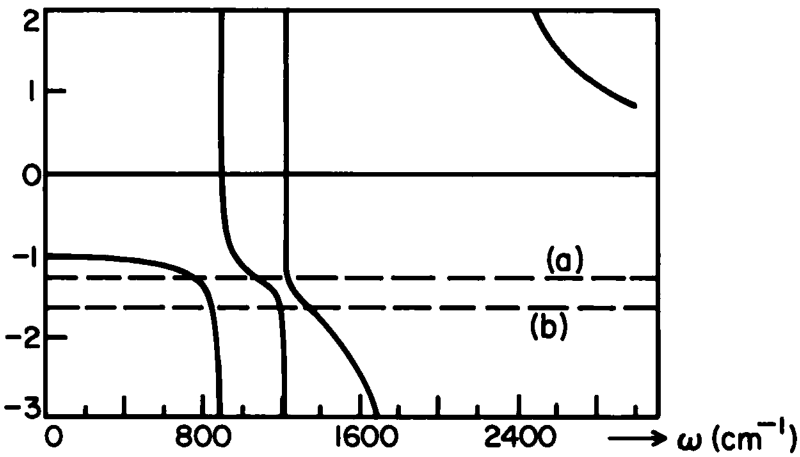

FIGURE 1 The function $D_o(\omega)$ (Eq. 13) with the parameters of Table II. The intersections with the value $-1/(1-\alpha)$ = -1.26 (line a) give the IR frequencies ω_n^{ϕ}, while the intersections with the value $-1/(1-2\lambda)$ = -1.62 (line b) give the Raman frequencies ω_n^R.

Some properties of the conductivity Eq. (12) are worth discussing. First note that the information on the nature of the charge, i.e. if it is a soliton, a polaron[15], a soliton lattice[16] or any other configuration, is contained in the single parameter M_c. In fact, the conductivity in the incommensurate limit (or high soliton density) has been calculated[12,13] and it coincides with Eq. (12) when $(\omega/\Delta_o)^2 \ll 1$, ρ is the total charge and $M_c = M_F$ the Fröhlich mass. Thus the IR frequencies, as well as their relative weights, are independent of the charge configuration, as long as $(\omega_n^{\phi}/\Delta_o)^2 \ll 1$. The value of M_c can be derived by comparing the IR intensities with the total intensity which includes electronic interband transitions. For this comparison one needs the conductivity sum rule[19]

$$\int_0^{\infty} \mathrm{Re}\sigma(\omega)\,d\omega = e^2 v_F \ . \tag{14}$$

Another possibility for determining M_c is by measuring the dielectric constant

$$\varepsilon(0) = 1+4\pi\rho e^2/(\alpha M_c \Omega_o^2) \ . \tag{15}$$

The term 1 in Eq. (15) is different in reality because of inner core polarizability, however, the term linear with should yield the value of M_c.

Another feature of Eq. (12) is a "product rule" of the frequencies ω_n^ϕ. The denominator in Eq. (12) can be written as $\prod_n(\omega^2-\omega_n^{\phi 2})/(\omega^2-\omega_n^{o 2})$. By comparing values at $\omega=0$ the product rule is obtained

$$\prod_{n=1}^{N} (\omega_n^\phi/\omega_n^o)^2 = \alpha \ . \tag{16}$$

To derive the parameters ω_n^o, λ_n, consider next the Raman frequencies of the dimerized (undoped) system,[13,17] which correspond to amplitude oscillations around Δ_o.

If $-N(0)E_i(\Delta)$ is the interaction energy of forming a gap $\Delta=\sum_n\Delta_n$ and $N(0)=2/\pi v_F$, then the effective Lagrangian is

$$L_{eff}\{\Delta\} = N(0)\left\{\sum_n \frac{1}{4\lambda_n} -\Delta_n^2+(\dot{\Delta}_n/\omega_n^o)^2 +E_i(\Delta)\right\} \ . \tag{17}$$

$E_i(\Delta)$ is independent of $\dot{\Delta}$ since the dynamics are dominated by the phonon terms. This is the same adiabatic principle used above which is valid for $\omega<<\Delta_o$.

The ground state is $\Delta_o = 2\lambda E_i'(\Delta_o)$ while small oscillations with amplitude $\delta_n(t)$ satisfy

$$\delta_n(t)+\ddot{\delta}_n(t)/\omega_n^{o2} = 2\lambda_n\delta(t)E_i''(\Delta_o) \tag{18}$$

where $\delta = \sum_n\delta_n$. The eigenfrequencies of Eq. (18) solve the equation

$$D_o(\omega) = -1/(1-2\tilde{\lambda}) \tag{19}$$

where $D_o(\omega)$ was defined in Eq. (13) and $1-2\tilde{\lambda} = 2\lambda E_i''(\Delta_o)$.

The interaction energy $E_i(\Delta)$ depends on both electron-phonon and electron-electron interactions. For the Peierls model (no electron-electron interactions)

$E_i(\Delta) = \frac{1}{4}\Delta^2 + \frac{1}{2}\Delta^2\ell n(2E_c/\Delta)$ [18] where E_c is the electron cutoff energy. Thus $\Delta_o = 2E_c\exp(-1/2\lambda)$ and in Eq. (19) $\lambda=\tilde{\lambda}$. This coincides with the Raman frequencies in the incommensurate limit[13] except that λ is replaced by $2\tilde{\lambda}$.[11]

Eq. (19) is identical to the equation for ω_n^ϕ except α is replaced by $2\tilde{\lambda}$. Therefore it has N solutions for the Raman frequencies ω_n^R; for $2\tilde{\lambda}<1$, $\omega_1^R<\omega_1^o$ and there is one additional solution in each interval $(\omega_n^o, \omega_{n+1}^o)$ (see Fig. 1).

Following the derivation of Eq. (16), the Raman frequencies ω_n^R satisfy the product rule

$$\prod_{n=1}^{N} (\omega_n^R/\omega_n^o)^2 = 2\tilde{\lambda} \ . \tag{20}$$

The ratio of Eqs. (16) and (20) gives

$$\prod_{n=1}^{N} (\omega_n^\phi/\omega_n^R)^2 = \alpha/2\tilde{\lambda} \ . \tag{21}$$

The significance of this result is that the left hand side involves measurable data while the right hand side is isotope independent since α and $\tilde{\lambda}$ involve only electronic properties.

It should be emphasized that the product rule Eq. (21) as well as the conductivity Eq. (12) are of general validity - the effects of electron-electron interactions are contained in the parameters M_c and $\tilde{\lambda}$.

From the data on $(CH)_x$ and $(CD)_x$ (see Tables I, II) the left hand side of Eq. (21) is 0.61 for $(CH)_x$ and 0.55 for $(CD)_x$. In view of the experimental uncertainty (ω_1 has a width of $\sim$400 cm^{-1}) these numbers are consistent with each other. Considering the large frequency shifts between $(CH)_x$ and $(CD)_x$ and the change in the number N of modes the result that the product rule remains unchanged is quite remarkable.

Consider now the Peierls model ($\tilde{\lambda}=\lambda$); from data on $\Delta_o/2E_c$, λ is obtained and the IR and Raman frequencies yield 2N equation for the 2N unknowns ω_n^o, λ_n/λ and α. For polyacetylene $2E_c$ = 10 eV, Δ_o = 0.7 eV [6] so that λ = 0.19. Note also that in resonance Raman scattering the intensity of phonons which are coupled to the extended π electrons is strongly enhanced. Thus in $(CH)_x$ there are two coupled modes while in $(CD)_x$ there are three modes.[20-22] The IR data shows indeed two ω^ϕ modes in $(CH)_x$ [2] and three ω^ϕ modes in $(CD)_x$.[5] The three modes in $(CD)_x$ appear, however, only upon doping with Na. Doping $(CD)_x$ with AsF_5 or I_2 shows[3-5] only two modes, but the higher frequency mode is much wider than ω_2^ϕ of $(CH)_x$ and should therefore be considered as two overlapping IR modes.

Tables I and II summarize the experimental data; the IR modes of $(CD)_x$ are those with Na doping.[5] Using λ=0.19 the values of ω_n^o and λ_n/λ are obtained, as shown in the tables. Also shown are the weights W_i of the IR modes relative to

TABLE I Parameters of trans $(CH)_x$: IR(ω_n^{ϕ}), Raman (ω_n^R) and bare (ω_n^o) frequencies in cm^{-1}, coupling constants λ_n, ($\lambda = \sum_n \lambda_n = 0.19$) and relative weight W_n in the sum rule (Eq. 14) in units of $\rho/M_c v_F$.

ω_n^{ϕ} Refs 2,5	ω_n^R Refs 20,21	ω_n^o	λ_n/λ	W_n
900	1075	1210	.08	1.2
1370	1470	2110	.92	0.5

TABLE II Parameters of trans $(CD)_x$. Notations as in Table I.

ω_n^{ϕ} Ref 5	ω_n^R Refs 20,22	ω_n^o	λ_n/λ	W_n
760	850	890	.04	1.6
1070	1200	1220	.007	2.0
1240	1340	2040	.953	0.3

the conductivity sum rule (Eq. 14) in units of $\rho/M_c v_F$. The intensity ratio is in reasonable agreement with experimental estimate[4] $W_1/W_2 \simeq 2$ for $(CH)_x$ and $W_1/(W_2+W_3) \simeq 0.7$ for $(CD)_x$.

The weight W_n of a particular mode is affected by the coupling of all higher frequency modes, and not just by its own coupling λ_n/λ. Thus in $(CH)_x \lambda_1/\lambda_2 \simeq 0.1$ but $W_1/W_2 \simeq 2.2$ while in $(CD)_x$ the mode with $\lambda_2/\lambda = .007$ has the strongest intensity. Note also that although the pinning force α is isotope independent, the pinned frequency ω_1^{ϕ} is isotope dependent as it is determined by the balance of all masses in the system.

The mean frequency of Eq. (6) is Ω_o=1960 cm^{-1} for $(CH)_x$

and Ω_o=1880 cm^{-1} for $(CD)_x$. The corresponding soliton masses are M_s/m_e=2.7;2.9.

The distribution of the dopant ions leads to a distribution $P(\alpha)$ of pinning parameters α; the observed absorption is then $\bar{\sigma}(\omega) = \int \sigma(\omega)P(\omega)d\omega$. Assuming a Gaussian distribution with width of $\Delta\alpha$=0.1 the absorption for $(CH)_x$ is shown in Fig. 2.

The result is in good agreement with experimental data.[2,4] Note that the distribution α affects mainly the lowest frequency, i.e. the "pinned mode".

Mele and Rice[7] claimed that there is a third mode in $(CH)_x$ well below 900 cm^{-1} which is the pinned mode. This cannot be the case since there is only one mode below ω_1^o and 900 cm^{-1} < ω_1^R < ω_1^o , i.e. the 900 cm^{-1} is the pinned mode in view of the Raman data. The results of Mele and Rice[7] may be understood in terms of lattice discreteness - a weak lattice pinning[8] of a light soliton may lead to a rather high pinning frequency. This effect is irrelevant to the experiment since quantum and thermal fluctuations can easily overcome the low binding energy of $\sim$16 cm^{-1}.[8] The relevant pinning is due to the dopants ions as considered above.

Finally I consider various methods for determining the mass M_c from experiment. The pinning parameter α determines M_c if the pinning potential and the charge distribution were known; however these functions are not reliably known.

The mass M_c can also be determined by the dependence of $\varepsilon(0)$ on ρ, Eq. (15). However the experiments are not conclusive[23,24] and it is also not clear how to relate $\varepsilon(0)$

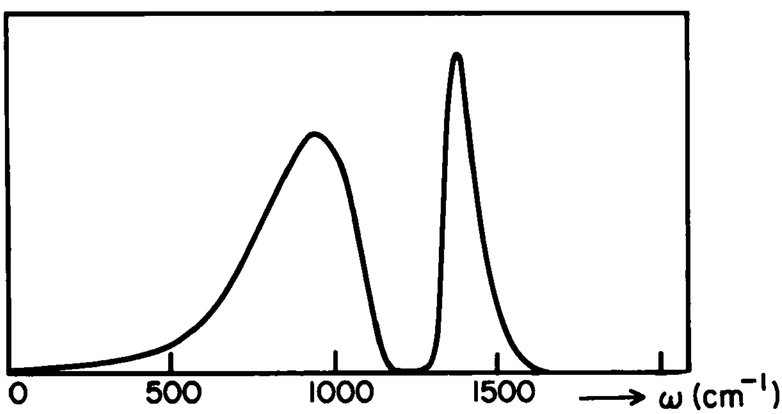

FIGURE 2 Absorption of doped $(CH)_x$ (parameters from Table I) with a Gaussian distribution for α with width 0.1 around α=0.2.

along the chain to the measured bulk dielectric constant.

The most reliable method of determining M_c is from the intensity ratio W_n/W_T where W_T is the total band absorption. Using[2,25] $W_2 = (2\text{-}3) \cdot 10^7\ \rho a\ cm^{-2}$ and $W_T = (1\text{-}4) \cdot 10^9\ cm^{-2}$ for $(CH)_x$ and the result in Table I, I obtain M_c/m_e= 15-100. This is considerably larger than the soliton or polaron masses. Possibly effects of electron-electron interactions and interchain coupling are needed to account for the experimental data.

In conclusion, I have shown the following results, within the adiabatic and continuum approximations:

a) The IR frequencies and the ratio of their intensities are independent of the charge configuration. Thus the claim[4,7] that the charged soliton configuration explains the IR data does not prove that solitons are indeed the charge carriers; the latter can be tested only if the electronic structure is involved.

b) The zero frequency translation mode acquires a finite frequency if pinning is present, e.g. due to Coulomb interaction with the dopant ion in doped polyacetylene. The number of IR modes, <u>including</u> the pinned mode, equals the number of Raman modes in the undoped system, or the number of bare coupled phonons. This confirms that the pinned mode in polyacetylene is at 900 cm^{-1} [2], and not at a much lower frequency as claimed in Ref. 7.

c) The product $\prod_n(\omega_n^\phi/\omega_n^R)$ is isotope independent (see Eq. 21). The product rule is in good agreement with data of $(CH)_x$ and $(CD)_x$. This remarkable result in fact justifies the neglect of lattice discreteness.

d) The results (a-c) are valid even if a direct electron-electron interaction is present. If, however, the latter is neglected, the parameters ω_n^o, λ_n and the pinning force can be determined, as summarized in Tables I, II. From the ratio of infrared to the total absorption intensity the mass M_c is estimated, and found to be considerably larger than the soliton or polaron masses. This suggests the importance of electron-electron interactions or interchain coupling.

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