

The cooperative transport of electrons and protons in the α -helix polypeptide chain

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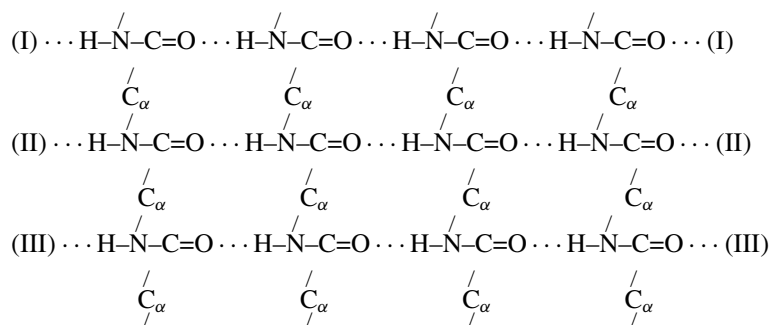
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Abstract. In this paper, we investigate the behaviour of electrons in the α -helix polypeptide chain and show that an electron can be bound in the compression area of the proton sublattice and become an electrosoliton state described by a bell-shaped electronic wave function. Due to the coupling between the three hydrogen-bonded chains, symmetric and asymmetric electrosoliton–soliton pairs are obtained.

1. Introduction

The charge transfer in hydrogen-bonded condensed matter, organic and biological systems is an important and interesting problem. A polypeptide chain with helix structure may consist of many hydrogen-bonded chains. For instance, an α -helical protein molecule composed of a single helical polypeptide chain has three hydrogen-bonded chains. The ‘melting’ of an α -helix structure can be performed only by the simultaneous breaking of a series of hydrogen bonds; it is a cooperative process. In other words, the maintenance of the helix structure of a polypeptide chain depends on the hydrogen bonds. In order to show the linking between three hydrogen-bonded chains we draw a schema of the α -helix protein molecule structure:



Obviously, interlinking between hydrogen-bonded chains depends on the α -carbon atom (C_α). In our previous paper [1], we investigated the proton transport in the α -helix polypeptide chain. In this paper, we investigate the charge transfer in the α -helix polypeptide chain.

As well know, many important properties of hydrogen-bonded system depend upon the motion of protons, such as the phase transition of hydrogen-bonded ferroelectrics and the

conductivity of ice. Solitonic defects are excited in the process of proton transfer, i.e. ionic and bonding defects are formed. The former involve an intrabond motion of the (unique) binding proton, while the latter result from interbond or intermolecule motion of the protons due to rotations of the molecules (e.g. the water molecules in ice). It has been shown that the ionic (I^-) and bonding (B^-) kink defects are carriers of a fractional negative charge so that their combined dynamics generates a 'proton hole' transfer $e_I^- + e_B^- = -e$, where $e > 0$ is the unit charge of one proton, to the next bond. On the other hand, the ionic (I^+) and bonding (B^+) antikink defects are carriers of a fractional positive charge, so that their combined dynamics generates a proton transfer ($e_I^+ + e_B^+ = e$) to the next bond. Antikinks excited in the proton sublattice possess excess fractional positive charge, corresponding to localized compression in the proton sublattice (for example, the hydroxonium ion H_3O^+ in an ice lattice), and kinks excited in the proton sublattice possess excess fractional negative charge, corresponding to localized rarefaction in the proton sublattice (for example, the hydroxyl ion OH^- in an ice lattice). In fact, the hydrogen-bonded chain can be considered to be composed of a proton sublattice and a heavy-ion sublattice. For example, the ice lattice is composed of a proton sublattice $(H^+)_x$ and a heavy-ion sublattice $(HO^-)_x$. Considering the influence of the motion of the heavy-ion sublattice on the proton sublattice, the component soliton model was suggested by some authors [2–6]. Because of the interaction between two sublattices, soliton defects corresponding to the heavy-ion sublattice localized deformation are excited [6]. On the other hand, there is a class of systems where the protonic and the electronic nature of the conductivity coexist at least for certain ranges of temperature [7, 8]. In biological systems, the coupling between proton and electron transfer is responsible for some reaction processes, for instance, there is the coupling between proton and electron transfer in charge relay system of α -chymotrypsin [9]. Thus, it is necessary to research the behaviour of the electrons, i.e. investigate the influence of the deformation of proton and heavy-ion sublattices upon the motion of electrons. (Abdullaev *et al* investigated only the system of protons and electrons [10].) The cooperative transport of electrons and protons in a simplex hydrogen-bonded chain was discussed in our previous paper [11]. This paper develops from the simplex chain theory into the three-chain theory. For the sake of simplicity, we do not alone consider the interaction between electron and heavy-ion sublattices but incorporate it into the coupling between protons and electrons because interaction between proton and heavy-ion sublattices is considered. The results in this paper show that one electron can be trapped by the lattice defects (slow antikink–kink pair or fast antikink–antikink pair) in the hydrogen-bonded chain. The bell-shaped electronic wave function (electrosoliton state) is localized in the compression area of the proton sublattice. The lattice defects and electrosoliton state propagate along the hydrogen-bonded chain with the same velocity in pairs, representing a bound state called an electrosoliton–soliton pair, which possesses a fractional negative charge [12]. Moreover, due to the coupling between the three hydrogen-bonded chains, symmetric and asymmetric electrosoliton–soliton pairs are obtained.

2. Hamiltonian and equations of motion

Our theory for three hydrogen-bonded chains develops from the simplex chain theory. We assume that the coupling between the hydrogen-bonded chains is a linear interaction, and consider the interaction between electron and lattice defects in the hydrogen-bonded chains. The total Hamiltonian of the system is

$$H_{tot} = H_{pe} + H_{ph} + H_h + H_i \quad (1)$$

where

$$H_{pe} = \sum_{n,\sigma} \left[\frac{1}{2} m_1 \left(\frac{du_{n,\sigma}}{dt} \right)^2 + \frac{1}{2} m_1 \left(\frac{c_0}{a} \right)^2 (u_{n+1,\sigma} - u_{n,\sigma})^2 + \varepsilon_0 \left(1 - \frac{u_{n,\sigma}^2}{u_0^2} \right)^2 \right. \\ \left. + E_0 C_{n\sigma}^+ C_{n\sigma} - J (C_{n+1,\sigma}^+ C_{n\sigma} + C_{n\sigma}^+ C_{n+1,\sigma}) + \chi (u_{n\sigma} - u_{n-1,\sigma}) C_{n\sigma}^+ C_{n\sigma} \right] \quad (2)$$

is the Hamiltonian of the proton–electron system [12],

$$H_{ph} = g_1 \sum_{n,\sigma} (u_{n,\sigma} - u_{n-1,\sigma})(\rho_{n,\sigma} - \rho_{n-1,\sigma}) \quad (3)$$

is the Hamiltonian for interaction between proton and heavy-ion sublattices [13],

$$H_h = \sum_{n,\sigma} \left[\frac{1}{2} m_2 \left(\frac{d\rho_{n,\sigma}}{dt} \right)^2 + \frac{1}{2} m_2 \left(\frac{v_0}{a} \right)^2 (\rho_{n+1,\sigma} - \rho_{n,\sigma})^2 \right] \quad (4)$$

is the Hamiltonian of the heavy-ion sublattice [6] and

$$H_i = g_2 \sum_{n,\sigma} (\rho_{n,\sigma} - \rho_{n-1,\sigma})(\rho_{n,\sigma+1} - \rho_{n-1,\sigma+1} + \rho_{n,\sigma-1} - \rho_{n-1,\sigma-1}) \quad (5)$$

is the Hamiltonian for interaction between hydrogen-bonded chains. a is the lattice spacing. $\sigma (= 1, 2, 3)$ is the sign of the chain. c_0 and v_0 are the characteristic velocities of the proton and heavy-ion sublattices, respectively. ε_0 is the barrier height in the double-well potential. $u_{n,\sigma}$ is the displacement of the n th proton (mass m_1) in the σ th chain along the chain from one of the two minima in the double-well potential. $\rho_{n,\sigma}$ is the displacement of n th heavy ion (mass m_2) in the σ th chain from its equilibrium position. u_0 is the equilibrium position of the proton. g_1 is the coupling constant between proton and heavy-ion sublattices in the σ th chain. g_2 is the coupling between hydrogen-bonded chains. E_0 is the energy of the electron in the undistorted chain, J is the intersite transfer energy and $\chi > 0$ is the coupling constant of the interaction between the electronic and the protonic subsystems. Finally, $C_{n\sigma}^+$ ($C_{n\sigma}$) creates (annihilates) one electron on the n th site of the σ th protonic chain. The same Hamiltonian can be used in order to describe the dynamics of one electron hole in the chain. In this case, $\chi < 0$, and $C_{n\sigma}^+$ ($C_{n\sigma}$) creates (annihilates) one hole on the n th site of the σ th chain.

The one-electron state can now be written [12]

$$|\Psi_\sigma(t)\rangle = \sum_n A_{n\sigma}(t) C_{n\sigma}^+ |0\rangle \quad (6)$$

where the probability amplitude is normalized to unity i.e. $\sum_n |A_{n\sigma}(t)|^2 = 1$. The mean value of the Hamiltonian in the system is written

$$E = \sum_{n,\sigma} A_{n,\sigma}^* [E_0 A_{n\sigma} - J (A_{n+1,\sigma} + A_{n-1,\sigma}) + \chi (u_{n,\sigma} - u_{n-1,\sigma}) A_{n\sigma}] \\ + \sum_{n,\sigma} \left[\frac{1}{2} m_1 \left(\frac{du_{n,\sigma}}{dt} \right)^2 + \frac{1}{2} m_1 \left(\frac{c_0}{a} \right)^2 (u_{n+1,\sigma} - u_{n,\sigma})^2 + \varepsilon_0 \left(1 - \frac{u_{n,\sigma}^2}{u_0^2} \right)^2 \right] \\ + \sum_{n,\sigma} \left[\frac{1}{2} m_2 \left(\frac{d\rho_{n,\sigma}}{dt} \right)^2 + \frac{1}{2} m_2 \left(\frac{v_0}{a} \right)^2 (\rho_{n+1,\sigma} - \rho_{n,\sigma})^2 \right] \\ + \sum_{n,\sigma} g_1 (u_{n,\sigma} - u_{n-1,\sigma})(\rho_{n,\sigma} - \rho_{n-1,\sigma}) \\ + \sum_{n,\sigma} g_2 (\rho_{n,\sigma} - \rho_{n-1,\sigma})(\rho_{n,\sigma+1} - \rho_{n-1,\sigma+1} + \rho_{n,\sigma-1} - \rho_{n-1,\sigma-1}). \quad (7)$$

In the continuum approximation model, from the Lagrangian density we can derive the equations of motion

$$i\hbar \frac{\partial A_\sigma}{\partial t} = (E_0 - 2J)A_\sigma - Ja^2 \frac{\partial^2 A_\sigma}{\partial x^2} + \chi a \frac{\partial u_\sigma}{\partial x} A_\sigma \quad (8)$$

$$m_1 \frac{\partial^2 u_\sigma}{\partial t^2} = m_1 c_0^2 \frac{\partial^2 u_\sigma}{\partial x^2} + 4\varepsilon_0 \frac{u_\sigma}{u_0^2} \left(1 - \frac{u_\sigma^2}{u_0^2}\right) + \chi a \frac{\partial |A_\sigma|^2}{\partial x} + g_1 a^2 \frac{\partial^2 \rho_\sigma}{\partial x^2} \quad (9)$$

$$m_2 \frac{\partial^2 \rho_\sigma}{\partial t^2} = m_2 v_0^2 \frac{\partial^2 \rho_\sigma}{\partial x^2} + g_1 a^2 \frac{\partial^2 u_\sigma}{\partial x^2} + 2g_2 a^2 \left(\frac{\partial^2 \rho_{\sigma+1}}{\partial x^2} + \frac{\partial^2 \rho_{\sigma-1}}{\partial x^2} \right). \quad (10)$$

Obviously, equations (8)–(10) are coupled nonlinear equations.

3. Symmetric and asymmetric electrosoliton–soliton pairs

Under normal conditions, it is very difficult to obtain analytical solutions of equations (8)–(10). But, the analytic solutions are found with ease in the following two specific cases: (i) $\rho_1 = \rho_2 = \rho_3 = \rho$, $u_1 = u_2 = u_3 = u$; (ii) $\rho_1 = 0$, $\rho_2 = -\rho_3 = \rho$, $u_1 = u_0$, $u_2 = -u_3 = u$. Here we use a self-consistent method, and let self-consistent solutions of equations (8) and (9) have the following relationship

$$|A|^2 = q_1 u_x \quad (11)$$

where q_1 is an undetermined coefficient. Substituting equation (11) into equation (9) and using the variable transformation $\xi = x - vt$, we may reduce equations (9) and (10) to the following equation [6]:

$$\left(\frac{v^2}{c_0^2} - 1 - \frac{\chi a q_1}{m_1 c_0^2} - \frac{g_1 a^2 q_2}{m_1 c_0^2} \right) \frac{\partial^2 u}{\partial \xi^2} - \alpha u + \beta u^3 = 0 \quad (12)$$

where

$$\alpha = \frac{4\varepsilon_0}{m_1 c_0^2 u_0^2} \quad \beta = \frac{4\varepsilon_0}{m_1 c_0^2 u_0^4} \quad (13)$$

and

$$q_2 = q_{s2} = -\frac{g_1 a^2}{m_2(v_0^2 + 4g_2 a^2/m_2 - v^2)} \quad (14)$$

for case (i),

$$q_2 = q_{as2} = -\frac{g_1 a^2}{m_2(v_0^2 - 2g_2 a^2/m_2 - v^2)} \quad (15)$$

for case (ii). Integrating equation (12), we obtain

$$u = \delta u_0 \tanh \sqrt{\frac{\alpha}{2}} v(x - vt) \quad (16)$$

$$\rho = q_2 u \quad (17)$$

where

$$v = \left[1 + \frac{\chi a q_1}{m_1 c_0^2} + \frac{g_1 a^2 q_2}{m_1 c_0^2} - \frac{v^2}{c_0^2} \right]^{-1/2} \quad (18)$$

and $\delta = \pm 1$ is the polarity of the soliton. $\delta = 1$ corresponds to the kink solution, and $\delta = -1$ to the antikink. The solitons may be classified by their velocity into two modes as follows:

$$c_0^2 + \frac{\chi a q_1}{m_1} + \frac{g_1 a^2}{\sqrt{m_1 m_2}} > v^2 > v_0^2 + P \quad \text{fast mode} \quad (19)$$

$$v_0^2 + P - \frac{g_1 a^2}{\sqrt{m_1 m_2}} > v^2 > 0 \quad \text{slow mode} \quad (20)$$

where $P = P_s = 4g_2 a^2/m_2$ for case (i); $P = P_{as} = -2g_2 a^2/m_2$ for case (ii).

In an undeformed hydrogen-bonded chain ($\rho_1 = 0, u_1 = u_0$), the electron is described by the exciton state [14]. For a deformed hydrogen-bonded chain, substituting equation (11) into (8), we have

$$i\hbar \frac{\partial A}{\partial t} - (E_0 - 2J)A + Ja^2 \frac{\partial^2 A}{\partial x^2} - \frac{\chi a}{q_1} |A|^2 A = 0. \quad (21)$$

Equation (21) is a nonlinear Schrödinger equation, where $G = -\chi a/q_1$ is a coefficient of the nonlinear term of the NLS equation. If $G > 0$ equation (21) has an envelope-soliton solution, while for $G < 0$ equation (21) has a dark-soliton solution [15]. Here we are interested in the envelope-soliton solution, so taking $G > 0$ (i.e. $q_1 < 0$ for $\chi > 0$ or $q_1 > 0$ for $\chi < 0$), the solution of equation (21) is

$$A(x, t) = \left(\frac{G}{8J}\right)^{1/2} \text{sech} \left[\frac{G}{4Ja} (x - vt) \right] \exp[i(kx - \omega t)] \quad (22)$$

where

$$k = \frac{\hbar v}{2Ja^2} \quad \omega = \frac{E_0 - 2J + Ja^2 k^2 - G^2 (16J)^{-1}}{\hbar}. \quad (23)$$

Solutions $u(x, t)$ and $A(x, t)$ must satisfy self-consistent condition (11), so we have

$$\frac{G}{8J} \text{sech}^2 \frac{G}{4Ja} (x - vt) = -q_1 u_0 \sqrt{\frac{\alpha}{2}} v \text{sech}^2 \sqrt{\frac{\alpha}{2}} v (x - vt) \quad (24)$$

where $q_1 < 0$ (for $\chi > 0$), so taking the antikink solution ($\delta = -1$), corresponding to the compression area of the proton sublattice. From equation (24), we obtain

$$q_1 = -\frac{a}{2u_0} \quad (25)$$

$$v^2 = \frac{1}{2} \left[v_1^2 + v_0^2 + P \pm \sqrt{(v_1^2 - v_0^2 - P)^2 + \frac{4g_1^2 a^4}{m_1 m_2}} \right] \quad (26)$$

where

$$v_1 = \left[1 - \frac{\chi a^2}{2m_1 u_0 c_0^2} - \frac{8J^2 a^2 \varepsilon_0}{m_1 u_0^4 c_0^2 \chi^2} \right]^{1/2} c_0 \quad (27)$$

is the velocity in the one-component soliton model of the simplex hydrogen-bonded chain. If $v_1^2 > v_0^2 + P$, v corresponds to the upper sign in equation (26) and $v^2 > v_0^2 + P$, i.e. $q_2 > 0$. Then, equations (14), (15) and (17) show that if the proton sublattice produces an antikink, the heavy-ion sublattice produces an antikink as well. They form a fast antikink–antikink pair. If $v_1^2 < v_0^2 + P$, v corresponds to the lower sign in equation (26) and $v^2 < v_0^2 + P$, i.e. $q_2 < 0$. Then, the antikink in the proton sublattice and the kink in the heavy-ion sublattice will form a slow antikink–kink pair. Here the antikink–antikink pair and antikink–kink pair are the lattice defects, i.e. the defects in proton and heavy-ion sublattices. These lattice defects correspond to

the compression area of the proton sublattice; thus, they have positive fractional extra charge [12, 16].

From the above discussion, it can be concluded that in a deformed hydrogen-bonded chain one electron is trapped by the lattice defects (slow antikink–kink pair or fast antikink–antikink pair) with a fractional positive charge to form a bound state with a fractional negative charge. The bell-shaped electronic wave function (electrosoliton state) is localized in the compression area of the proton sublattice. The lattice defects and electrosoliton state propagate along the hydrogen-bonded chain with the same velocity (given by equation (26)) in pairs, representing a bound state called an electrosoliton–soliton pair (or radical electron–soliton state [12]). However, under case (ii), if there is an antikink in one of two deformed hydrogen-bonded chains, then there is a kink in the other deformed chain. Corresponding to the kink solution ($\delta = 1$), taking $q_1 > 0$ (for $\chi < 0$), equation (25) becomes $q_1 = a/2u_0$. Using the same method, we can obtain the dynamics of one electron hole in the chain, where a bell-shaped hole wave function is localized in the rarefaction area of the proton sublattice. One hole is trapped by another kind of lattice defect (slow kink–antikink pair or fast kink–kink pair) with a fractional negative charge to form a bound state with a fractional positive charge. On the other hand, in an undeformed hydrogen-bonded chain, the electron is described by the exciton state wave function. The validity of the continuum approximation and stability of the electrosoliton–soliton pair have been discussed in detail in our previous paper [11].

The relative contraction of distance between neighbouring heavy ions in each chain can be given by

$$L = -\frac{\partial \rho}{\partial x} = -q_2 \delta u_0 \sqrt{\frac{\alpha}{2}} v \operatorname{sech}^2 \sqrt{\frac{\alpha}{2}} v (x - vt). \quad (28)$$

According to the discussion above, in case (i), three hydrogen-bonded chains produce local contraction (or expansion) deformation simultaneously. In case (ii), one hydrogen-bonded chain is undeformed, while if one of two deformed hydrogen-bonded chains is contracted, the other deformed chain is expanded. This leads to the local twisting deformation of the α -helix polypeptide chain. The electrosoliton–soliton pairs under case (i) and (ii) are called the symmetric and asymmetric electrosoliton–soliton pairs, respectively. The symmetric electrosoliton–soliton pairs, which contain three lattice defects and three electrosoliton states described by a bell-shaped electronic wave function, correspond to the local contraction (or expansion) deformation of the α -helix polypeptide chain. The asymmetric electrosoliton–soliton pairs, which contains one lattice defect and one electrosoliton state, and one other kind of lattice defect and hole state described by a bell-shaped hole wave function, correspond to the local twisting deformation of the α -helix polypeptide chain.

Using equation (7) and the continuum approximation model, we have the energy of the system

$$E = 3 \left(E_0 - 2J + \frac{\hbar^2 v^2}{4Ja^2} \right) + \lambda \left[(m_1 + q_2^2 m_2) v^2 - \frac{u_0^2 \chi^2}{4J\Delta} + (m_1 c_0^2 + q_2^2 m_2 v_0^2) + \frac{4\varepsilon_0}{3av\Delta} \sqrt{\frac{2}{\alpha}} + (g_1 + \eta) q_2 \right] \quad (29)$$

where $E = E_s$, $q_2 = q_{s2}$, $\lambda = 3\Delta$, $\eta = g_2 q_{s2}$ for case (i); $E = E_{as}$, $q_2 = q_{as2}$, $\lambda = 2\Delta$, $\eta = -\frac{1}{2} g_2 q_{as2}$ for case (ii) and $\Delta = (2vu_0^2/3a)\sqrt{\alpha/2}$. Therefore, it is evident that $E_s > E_{as}$. Namely, under case (ii), the coupling between chains decreases the energy of the system. Thus, the system is more stable than under case (i). On the other hand, it is shown from equation (29) that the binding energy of the electrosoliton–soliton pair is $u_0^2 \chi^2 / 4J$, that is, the internal energy

of the bound state is lower than the sum of the internal energy of the free lattice defects and free exciton.

From the above discussion, it follows that the interaction between the three hydrogen-bonded chains is responsible for the existence of a symmetric and asymmetric electrosoliton–soliton pair, which represents a bound state which a fractional negative charge. In order to justify the physical interest of our model, we choose the experimental parameters as follows [4, 14]: $a = 2.76 \text{ \AA}$, $u_0 = 0.37 \text{ \AA}$, $c_0 = 1.1 \times 10^5 \text{ m s}^{-1}$, $v_0 = 0.1c_0$, $m_1 = 1.67 \times 10^{-27} \text{ kg}$, $m_2 = 17m_1$, $g_1 = 1.0 \times 10^{-10} \text{ kg s}^{-2}$, $g_2 = 1.0 \times 10^{-10} \text{ kg s}^{-2}$, $\varepsilon_0 = 0.67 \text{ eV}$, $J = 1.55 \times 10^{-22} \text{ J}$, $\chi = 1.0 \times 10^{-11} \text{ N}$. Substituting these parameters into equation (26), it can be shown that the influence of the interaction between the three hydrogen-bonded chains (i.e. the coupling constant g_2) on the velocity of electrosoliton–soliton pairs is so small that the velocities of symmetric and asymmetric electrosoliton–soliton pairs are about the same, i.e. $v \simeq 0.84 \times 10^5 \text{ m s}^{-1}$. The velocity is in the range of the fast mode (equation (19)). Therefore, according to the above discussion, fast-mode electrosoliton–soliton pairs are excited in α -helix polypeptide chains. Using above experimental parameters, $\Delta E = E_s - E_{as}$ can be obtained, i.e. $\Delta E \simeq 5.7 \times 10^{-19} \text{ J}$. The asymmetric electrosoliton–soliton pair is more stable and more easily excited than the symmetric. The binding energy of the electrosoliton–soliton pair is $2.2 \times 10^{-22} \text{ J}$.

In conclusion, due to interaction between the three hydrogen-bonded chain (i.e. g_2), the fast-mode asymmetric electrosoliton–soliton pair is responsible for the cooperative transport of electrons and protons in the α -helix polypeptide chain.

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