THE PROPERTIES OF RAMAN SCATTERING RESULTING FROM SOLITONS EXCITED IN THE ORGANIC PROTEIN MOLECULES

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Some properties of Raman scattering resulting from the solitons excited in the organic protein molecules have theoretically been studied in terms of the partial diagonalization of the soliton Hamiltonian. The transition probability and the differential cross-section of the Raman scattering are also given. The red-shift associating with the Stokes component of the Raman scattering obtained is found to be basically consistent with the experimental data in the systems.

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I. Introduction

We knew already that a new amide-I band (essentially C=O stretching) and a red shift of about 15 cm^{-1} from a main absorption peak at about 1665 cm^{-1} have recently been observed experimentally in infrared and Raman spectra of organic crystalline acetanilide, and protein molecules [1–8]. Obviously, this phenomenon is assigned to the existence and motion of solitons in the organic protein molecules, and this red shift is directly associated with the Stokes component of the Raman scattering corresponding to the energy gap between the soliton and exciton existed in the organic molecular crystals and protein molecules [1,2,8,9]. Therefore, to study the properties of Raman scattering of the solitons is helpful to understand and clarify essences and natures of the solitons excited in the organic molecular crystals and protein molecules. What is the purpose of this paper is to study the properties of Raman scattering resulting from the soliton motion, and to compare the theoretical result with the experimental datum. This paper is organized in the following manner. The theoretical analysis in Sec. II leads to dynamic equations of motion in the organic protein molecules and we find out the soliton solutions of the equations. In the Sec. III we make the partial diagonalization of

the soliton Hamiltonian and estimate the red shift of scattering lines resulting from the solitons. The transition probability and differential cross-section of Raman scattering are studied in Sec. IV.

II. The model Hamiltonian and the equations of motion of the solitons excited in the organic protein molecular

According to our results studied, the Hamiltonian describing the collective excitation resulting from the localized fluctuation of intramolecular excitation and deformation of structure of the molecular chains caused the energy released in ATP hydrolysis in the organic protein molecules is ever represented as [9–18]

$$H = H_{ex} + H_{ph} + H_{int},$$ (1)

where

$$H_{ex} = 1/2m \sum_n P_n^2 + m\chi_1^2/2 \sum_n r_n^2 + m\chi_2 \sum_n (R_{n+1} - R_{n-1})r_n^2 \qquad (2)$$

$$H_{ph} = 1/2M \sum_n P_n^2 + \beta/2 \sum_n r_n^2 - m\omega_1^2/2 \sum_n r_n r_{n+1} \qquad (3)$$

$$H_{int} = m\chi_1/2 \sum_n (R_{n+1} - R_{n-1})r_n^2 + m\chi_2 \sum_n (R_{n+1} - R_n)r_n r_{n+1}. \qquad (4)$$

They represent the properties of the intramolecular excitation caused by the localized fluctuation, and, the features of sound vibration of molecules caused by the deformation of structure of the molecular chains, and, the interaction between the two models of motion in the organic protein molecules, respectively. Where m is mass of amide-I vibrational quantum (exciton) generated by the intramolecular excitation in a peptide group, ω_0 and ω_1 are the diagonal and non-diagonal elements of dynamic matrix. ω_0 is also the Einstein resonant frequency, $\frac{1}{2}m\omega_1^2 r_n r_{n+1}$ is the interaction between the nearest neighbouring amide-I vibrational quanta in the molecular chain, r_n and $p_n = m\dot{r}_n$ are the normal coordinate of the nth vibrational quantum in the molecule and its canonical conjugate momentum, respectively; M is the mass of an amino acid molecule in an unit cell, $2\chi_1 = \partial\omega_0^2/\partial R$ and $2\chi_2 = \partial\omega_1^2/\partial R$ are the change of energy of vibrational quantum and of coupling interaction between the neighbouring vibrational quanta by unit extension of molecular chain, respectively; R_n and $P_n = M\dot{R}_n$ are the canonically conjugate operators of displacement and the momentum of the molecule ; β is the elastic constant of the molecular chains of the protein molecules.

Due to the fact that the intramolecular excitation and the vibration of molecular chains are all quantization, therefore we introduce the following canonical second quantization method: [9–18]

$$R_n = \sum_q (\hbar/2MN\omega_q)^{\frac{1}{2}} \frac{1}{\dot{a}_n}(a_q + a_{-q}^+)e^{inr_0q} ;$$

$$\tau_n = \sum_q (2m\omega_0/\hbar)^{-\frac{1}{2}} \frac{1}{\dot{b}_n}(b_n + b_n^+);$$

$$p_n = (\hbar m\omega_0/2)^{\frac{1}{2}} (b_n - b_n^+)(-i), \qquad (i = \sqrt{-1}) \qquad (5)$$

$$P_n = i\sum_q (N\hbar\omega_q/2N)^{\frac{1}{2}}(a_{-q}^+ - a_q)e^{inr_0q} \qquad (6)$$

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where $\omega_q = 2(\beta/M)^{\frac{1}{2}} \sin(r_0q/2)$ is the frequency of a phonon with wave vector q, N is the number of unite cells in the molecular chain, r_0 is the distance between the molecules, $b_n^+(b_n)$ and $a_q^+(a_q)$ are the creation (annihilation) operators of the exciton and phonon, respectively.

Utilizing Eqs. (5)–(6), Eqs. (1)–(4) become as

$$H = \sum_n \varepsilon_0(b_n^+ b_n + 1/2) - J\sum_n (b_n^+ b_{n+1} + b_n^+ b_{n-1}) + \sum_q \hbar\omega_q(a_q^+ a_q + 1/2)$$
$$+ \sum_{n,q} [g(q)(b_n^+ b_n + b_n b_n^+) + g_1(q)(b_n^+ b_{n+1} + b_n^+ b_{n+1}^+)](a_q + a_{-q}^+)\exp(inr_0q) \qquad (7)$$

where

$$g(q) = (\hbar/2MN\omega_q)^{1/2}(\hbar\chi_1/4\omega_0)[\exp(ir_0q) - \exp(-ir_0q)]$$

$$g_1(q) = (\hbar/2MN\omega_q)^{1/2}(\hbar\chi_2/2\omega_0)[\exp(ir_0q) - 1]; \quad \varepsilon_0 = \hbar\omega_0, \quad J = \hbar\omega_1^2/4\omega_0.$$

Utilizing the Heisenberg's equations of operator, and in the condition of exciton in the case of high density of excitons we here study (we here study not the properties of exciton in the case of high density), the equations of motion for the operators, b_n, a_q and a_{-q}^+ are, respectively, of the form

$$i\hbar\dot{a}_q = [a_q, H] = \hbar\omega_q a_q + \sum_n [g(q)(b_n^+ b_n + b_n b_n^+) + g_1(q)(b_n^+ b_{n+1} + b_n b_{n+1}^+)]e^{-inr_0q} \qquad (8)$$

$$i\hbar\dot{a}_{-q}^+ = [a_{-q}^+, H] = -\hbar\omega_q a_{-q}^+ - \sum_n [g_1(q)(b_n^+ b_n + b_n b_n^+) + g_1(q)(b_n^+ b_{n+1} + b_n b_{n+1}^+)]e^{-inr_0q} \qquad (10)$$

$$i\hbar\dot{b}_n = [b_n, H] = \varepsilon_0 b_n - J(b_{n+1} + b_{n-1}) + \sum_q [2g(q)b_n + g_1(q)(b_{n+1} + b_{n-1})](a_q + a_{-q}^+)e^{inr_0q} \qquad (9)$$

Due to the fact that the collective excitation generated by the localized fluctuation and deformation of structure has coherent feature in the organic protein molecules, therefore the trial wave function described this state in the system should ever be denoted in the following form [9–18]:

$$|\Phi\rangle = \frac{1}{\chi'}\left(1 + \sum_n \varphi_n(t)b_n^+\right)|0\rangle_{ex} \cdot U(t)|0\rangle_{ph} \qquad (11)$$

with

$$U(t) = \exp\left[\sum_n (1/i\hbar)(u_n(t)P_n - \pi_n(t)R_n)\right] \qquad (11a)$$

$$= \exp\left\{\sum_q (\alpha_q(t)a_q^+ - \alpha_q^*(t)a_q)\right\} \qquad (11b)$$

where $|0\rangle_{ex}$ and $|0\rangle_{ph}$ are the excitonic and phononic vacuum states, respectively. We here choose the normalized factor $\chi' = 1$ in the following calculation for convenience.

Using the formulae of expectation values of Heisenberg equations, Eqs. (8)-(10),

$$ih\frac{\partial}{\partial t}\langle\Phi|b_n|\Phi\rangle = \langle\Phi|[b_n, H]|\Phi\rangle; \qquad ih\frac{\partial}{\partial t}\langle\Phi|a_q|\Phi\rangle = \langle\Phi|[a_q, H]|\Phi\rangle \tag{12}$$

and Eq. (11), we can get the following equations of motion

$$ih\dot\varphi_n = \hbar\omega_0\varphi_n - J(\varphi_{n+1} + \varphi_{n-1}) + \sum_q [g(q)(a_q + a^*_{-q})\varphi_n$$

$$+ g_1(q)((a_q + a^*_{-q})\varphi_{n+1} + (a_q + a^*_{-q})\varphi_{n-1})]e^{inr_0q} \tag{13}$$

$$i\hbar\dot\alpha_q = \hbar\omega_q\alpha_q + \sum_n [2g(q)|\varphi_n(t)|^2 + g_1(q)(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]e^{-inr_0q} \tag{14}$$

$$i\hbar\dot\alpha^*_{-q} = -\hbar\omega_q\alpha^*_{-q} - \sum_n [2g(q)|\varphi_n(t)|^2 + g_1(q)(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]e^{-inr_0q} \tag{15}$$

From Eqs. (14)-(15), we can get

$$i(\dot\alpha_q + \dot\alpha^*_{-q}) = -\omega_q(\alpha_q - \alpha^*_{-q}) \quad \text{or} \quad (\ddot\alpha^*_{-q} + \ddot\alpha_q) = i\omega_q(\dot\alpha^*_{-q} - \dot\alpha_q)$$

$$i\hbar(\dot\alpha^*_{-q} - \dot\alpha_q) = -\hbar\omega_q(\alpha_q + \alpha^*_{-q}) - \sum_n [4g(q)|\varphi_n|^2 + 2g_1(q)(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]e^{-inr_0q}$$

Thus we can obtain from above two equations

$$(\ddot\alpha^*_{-q} + \ddot\alpha_q) = -\omega_q^2(\alpha_q + \alpha^*_{-q}) - \sum_n [4g(q)\omega_q/\hbar)(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]e^{-inr_0q}$$

$$+ (2g_1(q)\omega_q/\hbar)(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]e^{-inr_0q} \tag{16}$$

From Eq. (6) we may obtain the Fourier transformation, $u_n(t)$, of the variable u_q to be of the form

$$u_n(t) = N^{-\frac{1}{2}}\sum_q u_q(t)\exp(iqx), \quad (x = nr_0), \quad u_q(t) = (\hbar/2M\omega_q)^{\frac{1}{2}}(\alpha_q + \alpha^*_{-q}) \tag{17}$$

Substituting Eq. (17) into Eq. (16), we may obtain

$$\ddot u_q + \omega_q^2 u_q = \sum_n [(i\hbar\chi_1/(M\omega_0 N^{\frac{1}{2}}))\sin(r_0q)|\varphi_n|^2$$

$$+ (\hbar\chi_2/(2M\omega_0 N^{\frac{1}{2}}))(\exp(ir_0q)-1)(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]\exp(-inr_0q) \tag{18}$$

Eqs. (13) and (18) are just a set of differential equations of motion described the collective excitation and the collective motion. Very obviously, to find the solutions of the above equations is very difficult. However, if we study only the case of low frequency vibration of molecular chain, we may make use the long wave-length approximation,

i.e., $\sin(qr_0) \sim qr_0$, $(e^{ir_0q} - 1) \sim iqr_0$, and let $\omega_q = V_0q$, $V_0 = (\beta/M)^{\frac{1}{2}}r_0$ is the sound speed in the organic protein molecular chains. Thus Eq. (18) become as

$$\ddot u_q + V_0^2 q^2 u_q = N^{-\frac{1}{2}}\sum_q [(iq)(\hbar r_0\chi_1/(4M\omega_0))|\varphi_n|^2$$

$$+ (iq)(\hbar r_0\chi_2/(2M\omega_0))(\varphi^*_n\varphi_{n+1} + \varphi^*_n\varphi_{n-1})]e^{-inr_0q} \tag{19}$$

Now multiplying Eq. (19) with $N^{-\frac{1}{2}}\exp(inr_0q)$, and summing over the wave number, q, simultaneously, and again making the continuum approximation

$$\varphi_{n\pm1}(t) = \varphi_n(t) \pm r_0\frac{\partial}{\partial x}\varphi_n(t) + (r_0^2/2!)\frac{\partial^2}{\partial x^2}\varphi_n(t) + \cdots, \quad \varphi_n(t) \to \varphi(x, t)$$

$$u_{n\pm1}(t) = u_n(t) \pm r_0\frac{\partial}{\partial x}u_n(t) + (r_0^2/2!)\frac{\partial^2}{\partial x^2}u_n(t) + \cdots, \quad u_n(t) \to u(x, t) \tag{20}$$

and again utilizing Eq. (17) and

$$\frac{\partial^l}{\partial x^l}u(x, t) = N^{-\frac{1}{2}}\sum_q (iq)^l u_q(t)\exp(iqx), \quad (l = 1, 2, \cdots) \tag{21}$$

we can transform Eq. (19) into the following non-linear equation

$$\frac{\partial^2}{\partial t^2}u(x, t) - V_0^2\frac{\partial^2}{\partial x^2}u(x, t) = [\hbar r_0(\chi_1 + \chi_2)/M\omega_0]\frac{\partial}{\partial x}|\varphi(x, t)|^2 \tag{22}$$

At the same time, using Eqs. (20)-(21), then Eq. (13) becomes as

$$ih\frac{\partial}{\partial t}\varphi(x, t) = (\epsilon_0 - 2J)\varphi(x, t) - Jr_0^2\frac{\partial^2}{\partial x^2}\varphi(x, t) + (\hbar r_0(\chi_1 + \chi_2)/\omega_0)\varphi(x, t)\frac{\partial}{\partial x}u(x, t) \tag{23}$$

Eqs. (22)-(23) are a set of complete equations of motion of excitons and phonon in the organic protein molecules. Using now the boundary conditions:

$$\varphi(\pm\infty) = \varphi_x(\pm\infty) = u(\pm\infty) = u_x(\pm\infty) = 0 \tag{24}$$

and let $\zeta = x - vt$, thus the solution of Eq. (22) is of the form

$$\frac{\partial}{\partial x}u(x, t) = \frac{\partial}{\partial\zeta}u(x, t) = [\hbar r_0(\chi_1 + \chi_2)/(M\omega_0(V^2 - V_0^2))]|\varphi(x, t)|^2 \tag{25}$$

Inserting Eq. (25) into Eq. (23), we can get

$$ih\frac{\partial}{\partial t}\varphi(x, t) = (\epsilon_0 - 2J)\varphi(x, t) - Jr_0^2\frac{\partial^2}{\partial x^2}\varphi(x, t)$$

$$+ [\hbar^2 r_0^2(\chi_1 + \chi_2)^2/(M\omega_0^2(V^2 - V_0^2))]|\varphi(x, t)|^2\varphi(x, t) \tag{26}$$

The solution of soliton of Eq. (26) is of the form

$$\varphi(x, t) = (\mu/2)^{\frac{1}{2}}\text{sech}\left(\left(\frac{\mu}{r_0}\right)(x - x_0 - Vt)\right)\exp\{i[(\hbar V/2Jr_0^2)(x - x_0) - E_{sol}t/\hbar]\} \tag{27}$$

and

$$u(x,t) = \left[-\hbar(\chi_1 + \chi_2)/(2\omega_0\beta(1-S^2))\right] \tanh\left(\left(\frac{\mu}{r_0}\right)(x - x_0 - Vt)\right) \tag{28}$$

where

$$\mu = G/4J, \qquad G = \hbar^2 r_0^2(\chi_1+\chi_2)^2/(MV_0^2\omega_0^2(1-S^2)) \tag{29}$$

From Eq. (11b) and Eq. (11c), we may obtain

$$-i/\hbar \sum_n (u_n P_n - \pi_n R_n) = \sum_q (\alpha_q a_q^+ - \alpha_q^* a_q) \tag{30}$$

Utilizing Eqs. (6) and (17), we also can get

$$\alpha_q = (M\omega_q/2\hbar)^{\frac{1}{2}} u_q + i (1/2M\hbar\omega_q)^{\frac{1}{2}} \pi_q \tag{31}$$

$\pi_q = M\dot{u}_q$, at last, we can obtain

Again using Eqs. (6), (11), (17)-(18) and (17), we also can get

$$
\begin{aligned}
\alpha_q(t) &= N^{-\frac{1}{2}} \sum_n \alpha_n(t) e^{-iqx} \\
&= \sum_n \left[(M\omega_q/2\hbar N)^{\frac{1}{2}} u_n(t) + i (1/(2MN\omega_q))^{\frac{1}{2}} M\dot{u}_n(t)\right] \exp(-iqx) \\
&= i\hbar(\chi_1 + \chi_2)\pi M(\omega_q + Vq)/(4MV_0^2\omega_0(1-S^2)\hbar N\omega_q \mathrm{sh}(\pi r_0 q/2\mu)) e^{iqVt} \\
&= \alpha_q \exp(iqVt)
\end{aligned}
\tag{32}
$$

In such a case, the energy of the soliton can also be found out, ie.,

$$
\begin{aligned}
E_{sol} &= \int_{-\infty}^{\infty} H d\zeta \\
&= \int_{-\infty}^{\infty} \left[(\epsilon_0 - 2J)|\varphi|^2 + J r_0^2 \left|\frac{\partial \varphi}{\partial x}\right|^2 - G|\varphi|^4\right] d\zeta \\
&\quad + \frac{1}{2}\int_{-\infty}^{\infty} \left[M\left|\frac{\partial u}{\partial t}\right|^2 + \beta\left|\frac{\partial u}{\partial x}\right|^2\right] d\zeta \\
&= \epsilon_0 - 2J + \hbar^2 V^2/4Jr_0^2 - \mu^2 J/3 = E_0 + M_{sol}V^2/2
\end{aligned}
\tag{33}
$$

where $E_0 = \epsilon_0 - 2J - \hbar^4(\chi_1+\chi_2)^4/48\beta^2 J\omega_0^4$, $(S = V/V_0)$ is rest energy of the soliton.
The mass of the soliton is

$$M_{sol} = m + \hbar^4(\chi_1 + \chi_2)^2(1 - 3S^2/2 - S^4/2)/48\beta^2 J\omega_0^4 V_0^2(1 - S^2)^2 > m \tag{34}$$

Therefore, the energy and the rest energy of the soliton is small about $\hbar^4(\chi_1+\chi_2)^4/48\beta^2 J\omega_0^4(1-S^2)^2$ and $\hbar^4(\chi_1+\chi_2)^4/48\beta^2 J\omega_0^4$ than one of the exciton, $E = \epsilon_0 - 2J + mV^2/2$ and $E_0 = \epsilon_0 - 2J$, respectively, but the mass of the soliton is greater than one of the exciton, m. So, the soliton is in stable state.

III. Partial diagonalization of the soliton Hamiltonian in the system

In order to diagonalize partially the soliton Hamiltonian Eq. (7), we should naturally transform it to the intrinsic reference frame moving with the soliton at velocity V. In this case the soliton Hamiltonian Eq. (7) must be replaced first by

$$\bar{H} = H - \sum_k \hbar kV(b_k^+ b_k + a_k^+ a_k) \tag{35}$$

where

$$b_k^+ = N^{-\frac{1}{2}} \sum_n \exp(iknr_0)b_n^+, \qquad b_k = (b_k^+)^+ \tag{36}$$

Since the soliton excitation in the system is connected with the deformation of molecular chain or intramolecular distances, it is very necessary to pass in Eq. (35) to new phonons taking this deformation into account. Such a transition is realized by the replacement $R, P \to a_q^+, a_q \to A_q^+, A_q$ according to the following relations:

$$a_q^+ = \sum_n 1/\sqrt{2\hbar N}\left[(M\omega_q)^{\frac{1}{2}} R_n - i(M\omega_q)^{-\frac{1}{2}} P_n\right]\exp(inr_0q),$$

$$A_q^+ = a_q^+ - N^{-\frac{1}{2}}\alpha_q^*, \qquad A_q = a_q - N^{-\frac{1}{2}}\alpha_q \tag{37}$$

where $A^+(A)$ is the creation (annihilation) operator of the new phonon. The coherent phonon state (lattice distortion) then becomes as the vacuum state of the new phonons:

$$|\bar{0}\rangle_{ph} = \exp\left[(\alpha_q^+ a_q^+ - \alpha_q^* a_q)\right]|0\rangle_{ph} \tag{38}$$

where there is that

$$A_q|\bar{0}\rangle_{ph} = 0 \tag{39}$$

Now we carry out the canonical transformation for the partial diagonalization of the soliton Hamiltonian, i.e., we here introduce

$$B_\lambda^+ = \sum_n \varphi_\lambda(t)b_n^+; \quad B_\lambda = (B_\lambda^+)^+; \quad \sum_\lambda \varphi_\lambda^*(n)\varphi_\lambda(n') = \delta_{nn'}; \quad \sum_n \varphi_\lambda^*(n)\varphi_{\lambda'}(n) = \delta_{\lambda\lambda'} \tag{40}$$

The partial diagonalization of the Hamiltonian implies the diagonalization of that part of the Hamiltonian which does not contain the creation and annihilation operators of the new phonons, Eq. (37). The condition imposed onto the function $\varphi_\lambda(n)$ to realize such a diagonalization are equivalent in the continuum approximation to the following problem on eigenfunction $\varphi(n/\lambda)$ and the eigenvalues E_λ

$$\left[-J\frac{\partial^2}{\partial n^2} - (i\hbar V/r_0)\frac{\partial}{\partial n} - 2\mu^2 Jr_0^2 \mathrm{Sech}^2(\mu n) + \epsilon_0 - 2J\right]\varphi(n/\lambda) = E_\lambda \varphi(n/\lambda) \tag{41}$$

The equation (41) is analogous to Eq. (26). It has the unique bound state solution, which satisfies the boundary condition Eq. (24), to be

$$\varphi(n/s) = (\mu r_0/2)^{\frac{1}{2}} \mathrm{Sech}(\mu n) \exp(i\hbar Vn/(2Jr_0^2)) \tag{42}$$

with the energy

$$E_s = \varepsilon_0 - 2J - \hbar^2 V'^2/(4Jr_0^2) - \mu^2 J \qquad (43)$$

Simultaneously, it has also a unbound state solution, which does not satisfy the boundary condition Eq. (24), to be

$$\varphi(n/k) = \{[\mu r_0\, th(\mu n) - ikr_0]/(\sqrt{2\pi}(\mu - ikr_0))\}\exp[i(\hbar V/(2Jr_0^2) + kr_0)n] \qquad (44)$$

with the dispersion relation:

$$E_k = \varepsilon_0 - 2J - \hbar^2/(4Jr_0^2) + (kr_0)^2 J \qquad (45)$$

The E_s is less than that of the lowest unbound state $E_k^0 = \varepsilon_0 - 2J$ by the value $\mu^2 J$ in the statical state. The functions $\varphi(n/\lambda)$ are normalized as the following form

$$\int_{-\infty}^{\infty} dn\, \varphi^*(n/k)\varphi(n/k') = \delta(kr_0 - k'r_0);$$

$$\int_{-\infty}^{\infty} dn\, |\varphi(n/s)|^2 = 1; \qquad \int_{-\infty}^{\infty} dn\, \varphi^*(n/s)\varphi(n/k) = 0 \qquad (46)$$

Where B_k^+ creates an unbound exciton with the wave vector k, while B_s^+ create an excitation which is localized at the lattice distortion.

In obtaining Eq. (41) the parameter n was assumed to be continuous and the chain length to tend to infinity $N \to \infty$. Consequently, the wave vector k proved to be the continuous value which runs from $-\infty$ to ∞. In the following we mainly use a discrete description. The continuous description is transformed into a discrete one according to the relations:

$$\int_{-\infty}^{\infty} dn \to \sum_n, \qquad \int_{-\infty}^{\infty} dq \to 2\pi/(Nr_0)\sum_q, \qquad \delta(kr_0 - k'r_0) \to (N/2\pi)\delta_{kk'},$$

$$\varphi(n/s) \to \varphi_s(n), \qquad \varphi(n/k) \to (N/2\pi)^{\frac{1}{2}}\varphi_k(n) \qquad (47)$$

Thus, the Hamiltonian Eq. (35) being partially diagonalized becomes as [16,18]

$$\bar{H} = G^2/24J + E_s B_s^+ B_s + \sum_k E_k B_k^+ B_k + \sum_q \hbar(\omega_q - Vq)\alpha_q^+\alpha_q$$

$$+ (1/\sqrt{N})\sum_q \hbar(\omega_q - Vq)(1 - B_s^+ B_s)(A_q^+\alpha_q + A_q\alpha_q^*)$$

$$+ (1/\sqrt{N})\sum_{q,k} F(k,q)(B_{k+q}^+ B_k)(A_q^+ + A_{-q})$$

$$+ (1/N)\sum_{q,k} \bar{F}(k,q)(B_s^+ B_{-k} + B_k^+ B_s)(A_{-q}^+ + A_q) \qquad (48)$$

where

where

$$\bar{F}(k,q) = [2i\pi^4 q J\pi\hbar/(\omega_0(G + 4Ji\pi_0^3 k))](\hbar J/GN\omega_q)^{\frac{1}{2}}$$
$$\times [\chi_1(e^{iqr_0} - e^{-iqr_0}) + \chi_2(e^{iqr_0} - 1)]\, sech[2\pi r_0^3 J(q - k)/G]$$

$$F(k,q) = i(\hbar^2/(2N\omega_0^2\omega_q))^{\frac{1}{2}}[\chi_1(e^{iqr_0} - e^{-iqr_0}) + \chi_2(e^{iqr_0} - 1)]$$

$$\times \frac{[G^2 - 4iGJ(k+3q)r_0^3 + 16k(k+q)r_0^6 J^2]^{\frac{1}{2}}}{[G^2 - 4iJG(k+2q)r_0^3 + 16(k+q)r_0^6 J^2 k]} \qquad (49)$$

are the coupling constant of the bound state and the unbound state with the new phonons, respectively. Now notice that the distribution amplitude of the intramolecular excitation, Eqs. (42) and (44), are found if an exactly specified deformation potential is assumed to be given a priori. In this case, however, the bound state Eq. (42) unlike the unbound ones Eq. (44) is self-consistent with the deformation of chain. Such a self-consistent state of the intramolecular excitation and the deformation form a soliton which in the intrinsic reference frame is stationary.

Really

$$\bar{H}B_s^+|0\rangle_{ex}|0\rangle_{ph} = (E_s + G^2/24J)B_s^+|0\rangle_{ex}|0\rangle_{ph} \qquad (50)$$

From Eqs. (42)–(45) we see clearly that the soliton energy is less than that of the lowest excitonic state ($k = 0$) by the value of the binding energy, $\mu^2 J/3$. The exciton states unlike the soliton ones are delocalized. In general, at the given deformation in the molecular chain containing intramolecular excitations two types of states are possibly localized solitonic and delocalized excitonic states. The soliton is a dynamic self-sustaining entity resulting from a self-trapping of exciton interacting with the deformational molecular chain. Due to this self-trapping of the exciton, the energy of the system drops about $2\mu^2 J/3$ (It is just equal to the deformation energy of the molecular chain, $W = 1/N\sum_q \hbar(\omega_q - V_q)|\alpha_q|^2 = 2\mu^2 J/3$), thus, the energy of the soliton is less about $\mu^2 J/3$ than that of the lowest excitonic state, i.e., there is a gap between the solitonic and excitonic energy spectra with the value of $\mu^2 J/3$. Just so, a red shift of about 15 cm^{-1} from a main absorption speak at about 1665 cm^{-1} of amide I vibration in a peptide group in the organic protein molecular crystals appears in the Raman spectra. This red shift have been observed in the Raman scattering and Infrared experiments [1-8] for the crystalline acetainilide and the protein molecules, and so on. This energy, $\mu^2 J/3$, is the binding or forming energy of soliton. If utilizing the values of the physical parameters of the soliton and the gap. For example, $J = 1.55 \times 10^{-22}$ J; $\beta = (13 - 19.5)$ N/m; $\chi = \frac{\hbar_{x1}}{2\omega_0} = (20 - 62)$ PN, $\chi' = \frac{\hbar_{x2}}{2\omega_0} = (2 - 8)$ PN, for the protein molecules, we can, thus, $\omega_1 = (2 - 9) \times 10^{13}$ S^{-1}, $r_0 = (4 - 5)$ A for the protein molecules, we can find out the binding energy of the soliton to be about 12.73 cm^{-1}, which is basically consistent with above red shift value (15 cm^{-1}) of spectral lines observing in the Raman scattering experiment. This result shows that the red shift of the Raman spectral lines is resulted from the motion of the soliton in the systems. Meanwhile, this shows that the soliton is actually present in the organic protein molecules. In the follows we will calculate the transition probability of the Raman scattering.

IV. The transition probability and the differential cross-section of the Raman scattering resulting from the solitons

On the other hand, from above results we can see that identification of the corresponding Stokes component of the Raman scattering spectrum can prove to be one of the ways of detecting experimentally the energy gap between the soliton states and the exciton states and, hence, the solitons themselves. Hereupon, the calculation of differential cross-section of the Raman scattering resulting from the solitons is very necessary and reasonable. In this section we will estimate the size of differential cross-section arising from the motion of the solitons.

We here think the Raman scattering process to carry into effect via some interaction of molecular chains associated with the soliton, for example, electronic excitation whose Hamiltonian is

$$H_e = \sum_{km} \varepsilon_{m}(k) D_{km}^+ D_{km} \qquad (51)$$

where D_{km}^+ (D_{km}) is the creation (annihilation) operator of the electronic excitation of mth band with wave vector k and the energy $\varepsilon_m(k)$. At the same time, let the incident light wave be quantized in the volume $V' = N r_0 S'$ and denoted by the Hamiltonian

$$H_Q = \sum_{Q\sigma} \hbar \omega_Q C_{Q\sigma}^+ C_{Q\sigma} \qquad (52)$$

where $C_{Q\sigma}^+ (C_{Q\sigma})$ is the creation (annihilation) operator of the photon with the wave vector Q, the energy $\hbar\omega_Q$ and unit polarization vector $\vec{\varepsilon}_\sigma(Q)$.

According to the property of above partial diagonalized Hamiltonian, Eq. (48), the interaction Hamiltonian among the soliton, electronic excitation states and light wave leading to the Raman scattering may be represented as the following form

$$\begin{aligned}
\bar{H}_{int} &= 1/N \sum_{kk'q} \sum_{mm'} X_{mm'}(q) D_{k+qm}^+ D_{km'} b_{k'-q}^+ b_{k'} \\
&+ 1/N \sum_{kk'q} \sum_{mm'} \bar{X}_{mm'}(q) D_{km}^+ D_{k+qm'} b_{k'}^+ b_{k'-q} \\
&+ \sqrt{N} \sum_{Q\sigma m} U_{m\sigma}(Q)(C_{Q\sigma}^+ + C_{-Q\sigma})(D_{Q,m}^+ - D_{-Q,m})
\end{aligned} \qquad (53)$$

According to the property of above partial diagonalized Hamiltonian, Eq. (48), the

i.e., we here consider the interaction between the soliton state and the electronic excitation states, where $X_{mm'}$ and $\bar{X}_{mm'}$ are the interaction coefficients between the intramolecular excitations and the electronic excitations with different wave vectors, respectively, $U_{m\sigma}(Q)$ is the coupling constant of electronic excitations and the light wave, i.e.

$$U_{m\sigma}(Q) = -i\varepsilon_m(0)(2\pi/\hbar\omega_Q V)^{\frac{1}{2}} (\vec{\varepsilon}_\sigma(Q) \cdot \vec{d}_m) \qquad (54)$$

here $\vec{d}_m$ is the dipole moment of transition from the ground state of electron into state of mth band, and here Z-axis is directed along the molecular chain.

We now study the probability per unit time of the transition of the "light plus soliton (or exciton) and electron" system from the initial state to be of the form

$$|\beta\rangle = C_{Q_0\sigma_0}^+ |0\rangle_Q B_s^+ |0\rangle_{ex} \left(\prod_q (\bar{n}_q!)^{-\frac{1}{2}} (A_q^+)^{\bar{n}_q} \right) |\bar{0}\rangle_{ph} |0\rangle_e \qquad (55)$$

to the final state to be of the form

$$|f k\rangle = C_{Q\sigma}^+ |0\rangle_Q B_k^+ |0\rangle_{ex} \left(\prod_q (n_q!)^{-\frac{1}{2}} (a_q^+)^{n_q} \right) |\bar{0}\rangle_{ph} |0\rangle_e \qquad (56)$$

under the action of the perturbation potential, Eq. (53), where the initial state consisted of the photons with the wave vector Q_0 and polarization $\vec{\varepsilon}_{\sigma_0}(\vec{Q}_0)$ and the soliton moving with the velocity V and the new phonons and the electronic excitation, while the final state consisted of the photons with the wave vector Q and polarization $\vec{\varepsilon}_\sigma(Q)$ and the exciton (or a certain delocalized intramolecular excitation) (without solitons) and a certain number of ordinary phonons and electronic excitations. Where $|0\rangle_Q$ and $|0\rangle_e$ are the vacuum states of the photons and the electronic excitation states, respectively. Obviously, the representations of the initial state and the final state we here adopt have somewhat especial forms. According to the perturbation theory in quantum mechanics [21-22], in this case of interest the probability of transition from the initial state to the final state mentioned above can be determined by the expression

$$\begin{aligned}
\frac{d}{dt}\bar{W}(Q_0\sigma_0 - Q\sigma) &= \frac{d}{dt}\hbar^{-6} \sum_{k,ph} \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \\
&\times \langle fk|\bar{H}_{int}(t_1)\bar{H}_{int}(t_2)\bar{H}_{int}(t_3)|\beta\rangle |^2
\end{aligned} \qquad (57)$$

where $\bar{H}_{int}(t) = \exp(j H_0 t/\hbar)\bar{H}_{int}\exp(jH_0t/\hbar)$

$$H_0 = H + H_Q + H_e - \hbar \sum_{km} kVD_{km}^+ D_{km} - \hbar \sum_{Q\sigma} (\vec{D} \cdot \vec{V})C_{Q\sigma}^+ C_{Q\sigma} \qquad (58)$$

However, we ever are more interested in the long-time behavior of $d\bar{W}/dt$ for estimating this probability. By straightforward calculation the transition probability can approximately be represented as

$$\lim_{t\to\infty} \frac{d\bar{W}}{dt} = \frac{\pi r_0^3}{2\hbar^2\mu} \times$$

$$\left| \sum_{mm_0} \frac{(Q_{0z} - Q_z)U_{\sigma_0 m_0}(Q_0) \left[X_{mm_0}(Q_z - Q_{0z}) + \bar{X}_{mm_0}(Q_z - Q_{0z}) \right] U_{\sigma m}^*(Q)}{(\varepsilon_{m_0}(Q_{0z}) - \hbar\omega_{Q_0})(\varepsilon(Q_z) - \hbar\omega_Q)} \right.$$

$$\left. + \frac{(Q_z - Q_{0z})U_{\sigma_0 m_0}(Q_0)\left[X_{mm_0}(Q_z - Q_{0z}) + \bar{X}_{mm_0}(Q_z - Q_{0z}) \right]U_{\sigma m}^*(Q)}{(\varepsilon_{m_0}(-Q_{0z}) + \hbar\omega_{Q_0})(\varepsilon(-Q_z) + \hbar\omega_Q)} \right|^2$$

$$\cdot \operatorname{Re} \int_0^\infty dt \exp\left\{\frac{i}{\hbar}\left[E'_s - W - E_k + \hbar(\omega_0 - \omega_q) + \hbar(\bar{Q}_0 - \bar{Q}) \cdot \vec{V}\right] + g(t) + \zeta(t)\right\}$$

$$\cdot \int_{-\infty}^\infty dk \left[\left(\mu^2 + (k r_0^2)\operatorname{ch}^2\left(\frac{\pi r_0}{2\mu}(Q_{0z} - Q - K)\right)\right)\right]^{-1}$$ (59)

where

$$g(t) = 1/N \sum_q |f_q|^2 \left(\exp(-i(\omega_q - Vq)t) - 1\right);$$

$$\zeta(t) = -4/N \sum_q |f_q|^2 \sin((\omega_q - Vq)t/2);$$

$$E' = E_s + \varepsilon_0 - 2J - \hbar^2 V^2/4J r_0^2 - \mu^2 J$$ (60)

the off diagonal terms of the following matrix

$$U(kk'') = \langle f k'' | \exp\left(i \sum_q (\omega_q - Vq)A_q^+ A_q t\right) \cdot (A_k^+ + A_{-k})|\beta\rangle$$

$$\exp\left(-i \sum_q (\omega_q - Vq)a_q^+ a_q t\right) \cdot (A_{k''}^+ + A_{-k''})|\beta\rangle$$ (61)

are negligible unless $|k|$ and $|k''|$ are of the order of $2\mu/\pi r_0$ or less in above calculation. Since small wave vectors do not significantly contribute to the sum when $\pi^2/2\mu \gg 1$, we may replace $U(kk'')$ by $I(kk'') \delta_{kk''}$, where

$$I(kk'') = e^{i(\omega_k - kV)t}\langle f k''|A_k^+|A_k^+\rangle \exp\left(i \sum_q (\omega_q - Vq)A_q^+ A_q t\right)$$

$$\cdot \exp\left(-i \sum_q (\omega_q - Vq)a_q^+ a_q t\right) A_k|\beta\rangle$$

$$= \exp((\omega_k - kV)) \exp(g(t) + \zeta(t))$$ (62)

If the velocity of the soliton is small or $V \to 0$, thus we can approximately get $g(t) = g_0 \int_0^\infty x [\exp(-i\omega_a t x) - 1] dx/\sinh x$, where $g_0 = 2(\chi_1 + \chi_2)^2 \hbar/\omega_0^2 \omega_a V_0 \pi$, $\omega_a = 2\mu V_0/r_0\pi$. If $t > 0.002/\omega_a$, then $g(t)$ is well approximated by

$$g(t) = -g_0[\mathrm{Ln}(\omega_a t/2) + 1.587 + i\pi/2]$$

and

$$\zeta(t) = -4g_0 \int_0^\infty x \sin^2(\omega_a tx/2) \, dx/\sinh x = 2g_0 \left(1 - \pi\omega_a t/2 + \operatorname{cth}(\pi\omega_a t/2)\right)$$ (63)

we notice that

$$\lim_{t\to\infty} \zeta(t) = -\gamma t,$$

here $\gamma = g_0\pi\omega_a = \hbar^4(\chi_1 + \chi_2)^4/4\omega_0^4\beta^2 J\pi = 4\mu^2 J/\hbar\pi$ (64)

Now let

$$\Delta_{ks}(Q, Q) = E_k - E_s + \frac{2}{3}\mu^2 J + \hbar(\omega_q - \hbar\omega_{Q_0} - \hbar(\bar{Q} - \bar{Q}_0) \cdot \vec{V}$$ (65)

$$\Delta_{ks} = E_k - E_s + \frac{2}{3}\mu^2 J + \hbar\omega_q - \hbar\omega_{Q_0} = \Delta_{ks}(\omega_q, \omega_{Q_0})$$ (66)

for $V \to 0$. Utilizing above results of Eqs. (64)–(66) we may obtain

Since the solitonic velocity is small as compared with that of light, the last term in Eq. (65) can be neglected, thus we may get

$$\operatorname{Re} \int_0^\infty dt \exp[-\gamma t - g(t) - i(\Delta_{ks}(0) - W)t/\hbar]$$

$$= (2.42\,\omega_a)^{-g_0}\Gamma(1 - g_0)\left\{\gamma^2 + [(\Delta_{ks}(0) - W)/\hbar]^2\right\}^{\frac{g_0 - 1}{2}} \cos(g_0(\pi/2 - \varphi) + \varphi)$$

If using again above values of physical parameters for protein molecules, we can get that $g_0 = 0.006 - 0.011 < 1$, thus we can get

$$\operatorname{Re} \int_0^\infty dt \exp[-\gamma t - g(t) - i(\Delta_{ks}(0) - W)t/\hbar]$$

$$\approx (2.42\,\omega_a/\gamma)^{-g_0}(\hbar^2\gamma)/[\hbar^2\gamma^2 + (\Delta_{ks}(0) - W)^2]$$ (67)

Utilizing the above results obtained we can find out the integration over k in Eq. (59). As a result, for the transition probability, Eq. (59), we can get

$$\lim_{t\to\infty} \frac{d\overline{W}}{dt} = \frac{\pi^2 r_0^2}{4\hbar\mu} \times$$

$$\sum_{mm_0} \left| \frac{(Q_{0z} - Q_z)U_{\sigma 0 m_0}(Q_0)\left[X_{mm_0}(Q_z - Q_{0z}) + \bar{X}_{mm_0}(Q_z - Q_{0z})\right]U_{\sigma m}^*(Q)}{(\varepsilon_{m_0}(-Q_{0z}) - \hbar\omega_{Q_0})(\varepsilon(Q_z) - \hbar\omega_Q)} \right.$$

$$+ \frac{(Q_z - Q_{0z})U_{\sigma 0 m_0}(Q_0)\left[X_{mm_0}(Q_z - Q_{0z}) + \bar{X}_{mm_0}(Q_{0z} - Q_z)\right]U_{\sigma m}^*(Q)}{(\varepsilon_{m_0}(-Q_{0z}) + \hbar\omega_{Q_0})(\varepsilon(-Q_z) + \hbar\omega_Q)} \right|^2$$

$$\cdot \left\{ \operatorname{th}^2\left[\frac{\pi}{2\mu}\left((\Delta_{ks}/J)^{\frac{1}{2}} - (Q_{0z} - Q_z)r_0\right)\right] \right.$$

$$+ \operatorname{th}^2\left[\frac{\pi}{2\mu}\left((\Delta_{ks}/J)^{\frac{1}{2}} + (Q_{0z} - Q_z)r_0\right)\right] + 2 \right\}$$

$$\cdot \left\{ 2\left[(\Delta_{ks}(0) - W)^2 + (\hbar\gamma)^2\right]\left[\sqrt{(\Delta_{ks}(0) - W)^2 + (k\gamma)^2} - \sqrt{(\Delta_{ks}(0) - W)^2}\right]\right\}^{-\frac{1}{2}}$$

$$(\mu^2 J + \Delta_{ks})^{-1} \cdot (\hbar\gamma\sqrt{J}(2.42\,\omega_a/\gamma)^{-g_0})$$ (68)

According to quantum mechanics [21]-[22] the cross-section of Raman scattering is closely associated with the above transition probability, Eq. (68), thus we can find out the cross-section. Due to the fact that the transition probability, Eq. (68), has a sharp peak, when the frequency of scattering light in vicinity of incident light frequency ω_{Q_0}, therefore we are only of interest to the cross-section of Raman scattering in such a case. In fact, it is only worth to study the Raman scattering resulting from the expression for the differential cross-section of the Raman scattering resulting from the motion for the soliton in such a case for the organic protein molecules can be represented as

$$\frac{dN}{Nds} = \frac{d^2\delta}{d\omega_Q d\Omega} = \frac{V^2 \tilde{\omega}_Q^2}{4\pi^2}\frac{d\overline{W}}{dt} = \frac{r_0^2 \tilde{Q}_0^2 \omega_Q^2}{16\pi^3 JhC^6}\sum_\sigma$$

$$\left| \sum_{m m_0} \frac{(Q_{0z} - Q_z)U_{\sigma_0 m_0}(Q_0)\left[X_{m m_0}(Q_z - Q_{0z}) + \tilde{X}_{m m_0}(Q_z - Q_{0z})\right]U_{\sigma m}^*(Q)}{(\varepsilon_{m_0}(Q_{0z}) - \hbar\omega_{Q_0})(\varepsilon(Q_z) - \hbar\omega_Q)} \right.$$

$$+ \left. \frac{(Q_z - Q_{0z})U_{\sigma_0 m_0}(Q_0)\left[X_{m m_0}(Q_z - Q_{0z}) + \tilde{X}_{m m_0}(Q_{0z} - Q_z)\right]U_{\sigma m}^*(Q)}{(\varepsilon_{m_0}(-Q_{0z}) + \hbar\omega_{Q_0})(\varepsilon(-Q_z) + \hbar\omega_Q)} \right|^2$$

$$\cdot \left\{ \cos\theta_0 - \frac{\omega_0}{\omega_{Q_0}}(\sin\theta_0 \sin\theta \cos\varphi + \cos\theta_0 \cos\theta) \right\}^2 \left(\hbar\gamma\sqrt{J}(2.42\,\omega_a/\gamma)^{-g_0}\right)$$

$$\left\{ 2\left[(\Delta_{ks}(0) - W)^2 + (\hbar\gamma)^2\right]\left[\sqrt{(\Delta_{ks}(0) - W)^2 + (\hbar\gamma)^2} - \sqrt{(\Delta_{ks}(0) - W)^2}\right] \right\}^{-\frac{1}{2}}$$

$$\mathrm{sech}^2\left(\frac{\pi\omega_Q r_0}{2\mu c}\right)\left(\cos\theta_0 - \frac{\omega_Q}{\omega_{Q_0}}(\cos\theta_0 \cos\theta + \sin\theta_0 \sin\theta \cos\varphi)\right) \tag{69}$$

where $d\Omega = d\theta d\varphi \sin\theta$ is the element of solid angle in direction of the scattering light propagation, θ_0 is an angle between the incident light $\overline{Q}_0$ and Z-axis, θ is a angle between the $\overline{Q}_0$ and the Scattering light $\overline{Q}$, φ is a angle between the projection value of the vector $\overline{Q}$ on the XOY plane and X-axis. Obviously above the result of the differential cross-section of the Raman scattering resulting from the solitons has the following properties: (1) the frequencies of scattering light are very close to that of incident light; (2) the shapes of scattering spectral lines for a fixed direction of scattering are, in the main, decided by the function

$$\frac{\hbar\gamma\sqrt{J}(2.42\,\omega_a/\gamma)^{-g_0}}{\left\{2\left[(\Delta_{ks}(0) - W)^2 + (\hbar\gamma)^2 - (\Delta_{ks}(0) - W)\right]\left(\sqrt{(\Delta_{ks}(0) - W)^2 + (\hbar\gamma)^2} - (\Delta_{ks}(0) - W)\right)\right\}^{\frac{1}{4}}}$$

Also, the scattering indicatrices (angular dependence of the scattering cross-section) depends mainly on the orientation of the dipole moments of electronic transitions from the ground state to the resonance electronic levels in this case of $0 < \hbar\omega_{Q_0} < \mu^2 J - 2\varepsilon_n(0)$ or $(\mu^2 J + \varepsilon_n(0)) <$ but the scattering indicatrix in this case of $0 < \hbar\omega_{Q_0} < \mu^2 J < \hbar\omega_{Q_0} < \varepsilon_n(0)$, $\hbar\omega_{Q_0} < 2\varepsilon_0(0)$) is independent of coupling strength, when the frequency ω_Q of the

scattering light approach to that of the Raman peak; (3) the maximum of the scattering light is attained at the ω_Q of the order of $\omega_{Q_0} - \mu^2 J/3 + 2J - \varepsilon_0$, i.e., the most probable scattering in this case is that accompanied by the transition of the chain from the soliton state into the exciton state, when there are no deformations. Therefore, if we pay attention above the properties and remarkable asymmetry of the lines connected with the corresponding energy dependence and remarkable asymmetry of the density of delocalized states in hand (the long wave side of lines are smoother and the short-wave ones sharper) we can determine the existence of the soliton in the organic protein molecules from analysing the different properties of the Raman scattering spectra obtained experimentally.

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