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Influence of the soliton in anharmonic molecular crystals with temperature on Mössbauer effect

X.-F. Pang[a]

Institute of High-Energy Physics, University of Electronic Science and Technology of China, Chengdu 610054, P.R. China
Department of Physics, Kaiserslautern University, 67663 Kaiserslautern, Germany
International Centre for Material Physics, Academia Sinica, Shenyang 110015, P.R. China
Institute of Atom. and Mol. Physics, Sichuan University, Chengdu 610015, P.R. China

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Abstract. The influence of a soliton on the properties of the Mössbauer effect resulting from the gamma-active nuclei situated at lattice sites in quasi-one-dimensional anharmonically organic molecular chains, in which the active nuclei emit gamma-photons, has been theoretically studied by a quantum vibrational model. An expression for the gamma-radiated Mössbauer transition probability as a function of the temperature and non-linear interaction has been obtained. Finally we give some new properties of this effect from numerical calculation in such a case, which present a way to experimentally confirm the soliton existence in molecular crystals.

PACS. 76.80.+y Mössbauer effect; other γ-ray spectroscopy – 33.40.+f Multiple resonances (including double and higher-order resonance processes, such as double nuclear magnetic resonance, electron double resonance, and microwave optical double resonance) – 71.38.+i Polarons and electron-phonon interactions

1 Introduction

In the last thirty years it has been discovered that the non-linearity in the forces and soliton motion occurring in physical systems play a decisive role in the explanation of a large variety of phenomena. It was first of all found that the lattice was bound only by linear forces, *i.e.*, harmonic potentials which would have an infinite heat conductivity, and that only the introduction of non-linearity leads to finite vales of this quantity [1]. Perhaps the most interesting feature of non-linear dynamics is the existence of soliton solutions which have been observed in a lot of materials with non-dispersive and non-linear interactions, for example, water, ferro- and antiferro-magnets, optical fibre, and so on. Recently, there has been interest in the non-linear properties of the organic molecular crystals, for example, acetanilide $(\mathrm{CH_3COHNC_6H_5})_x$ or ACN [1–21]. In the acetanilide two close chains of hydrogen-bonded amide-I groups run through the crystal. The space group is $\mathrm{D}_{2\mathrm{h}}^{15}(\mathrm{P}_{\mathrm{bca}})$ and the unit cell of factor group is $\mathrm{D}_{2\mathrm{h}}$ for this kind of crystal. There are eight molecules in an unit cell and at the amide-I frequency each of these has one degree of freedom; there thus are three infrared-active modes ($\mathrm{B}_{1\mathrm{u}}$, $\mathrm{B}_{2\mathrm{u}}$, and $\mathrm{B}_{3\mathrm{u}}$), four Raman-active modes (A_g, $\mathrm{B}_{1\mathrm{g}}$, $\mathrm{B}_{2\mathrm{g}}$, and $\mathrm{B}_{3\mathrm{g}}$), and one inactive mode (A_u). However, at low frequency (< 200 cm^{-1}), each molecule exhibits six degrees of freedom (three of translation and three of rotation), thus there are a total of 48 low-frequency modes (24 Raman active modes ($6\mathrm{A}_\mathrm{g}+6\mathrm{B}_{1\mathrm{g}}+6\mathrm{B}_{2\mathrm{g}}+6\mathrm{B}_{3\mathrm{g}}$), 18 infrared-active modes ($6\mathrm{B}_{1\mathrm{u}}+6\mathrm{B}_{2\mathrm{u}}+6\mathrm{B}_{3\mathrm{u}}$) and six ($\mathrm{A}_\mathrm{u}$) modes corresponding to the acoustic modes of translation and rotation). All of these active modes are seen in infrared absorption and Raman experiments [1–5]. The ACN is an interesting system because the nearly-planar amide-I group display bond distances which are close to those found in polypeptides. Since the physical properties of such a hydrogen bonded amide-I system are very sensitive to bond distances, the study of ACN reveals some new phenomena [1–5]. For example, in the experiments of infrared absorption and Raman scattering, when the crystalline acetanilide is cooled below room temperature from 320 to 10 K, a new amide-I band red-shifted from the main peak, 1666 cm^{-1}, by about 16 cm^{-1}. No other major changes occur from 4000 cm^{-1} to 800 cm^{-1}. The intensity of this new band steadily increases from room temperature to 70 K. The band at 1650 cm^{-1} is not present in amorphous materials or ACN methylated at the position where the hydrogen-bonded distances occur, but after annealing it is fully recovered. Raman spectra of ACN show also the same results mentioned above [1–3].

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[a] e-mail: `pangxf@mail.sc.cninfo.net`

We knew that a characteristic feature of the amide-I group CONH in polypeptides is the amide-I mode, mainly involving C=O stretching. This mode is observed as an infrared absorption peak of about 1666 cm^{-1} in ACN and near that value in a wide variety of materials including the amide-I groups. The corresponding spectroscopic evidence of the new band have been presented, and detailed measurements of the crystal structure and specific heat as a function of temperature [1–3] preclude an assignment of the new band to (1) a conventional amide-I mode, (2) crystal defect states, (3) Fermi resonance or (4) frozen kinetics between two different subsystems. A correct idea is the assignment to a self-trapping of amide-I vibrational energy [1–5] on the basis of the following experimental facts: (1) ^{15}N substitution which induces a small shift on the amide-I at 1666 cm^{-1} also shifts the new band by the same amount; (2) deuterium substitution at the NH position strongly affects both the amide-I and the new band in a complicated way; (3) upon cooling a parallel decrease of the normal amide-I integrated absorption and increase of the 1650 cm^{-1} band integrated absorption is observed; (4) the 1650 cm^{-1} band and the amide-I band show the same dichroism over the temperature range integrated; (5) the measurements of specific heat, the dielectric constant and the volume expansion as a function of temperature, rule out the occurrence of rotational isomerism or of a polymorphic transition which would affect some other infrared and Raman absorption bands, but not affect the new band. The self-trapping mechanism of amide-I vibrational energy proposed by Scott, Eilbeck *et al.* [1–5] comes from the Davydov model of vibrational energy transport in alpha-helix protein [9]. They have given a good account of the properties of first excited state in this model. Scott *et al.* [4] and Alexander *et al.* [6] have also obtained the exponential dependence of the absorption intensity on the temperature, $\exp(-\beta T^2)$, and explain the changes of intensity of the new band with decreasing temperature observed in the experiments [1–6] in terms of a complementary polaron or soliton pictures of the self-trapping state on the basis of the Davydov model. However, the red-shifted value obtained from these models is too small, only a few cm^{-1} as compared with the experimental value of 16 cm^{-1}. This shows these models need improving.

We here think, also, that the new band of amide-I is brought about by the self-trapping of amide-I vibrational quanta [12–18] which is different from the Davydov's exciton-soliton model or Alexander's complementary polaron picture. We think that the cooperative interaction between the localized amide-I vibrations and low frequency lattice vibrations results in the occurrence of the vibron-soliton, *i.e.*, thought such an intrinsically non-linear interaction between them, an amide-I vibrational quantum as a source of the low-frequency phonons causes shifts in the average positions of the ground state of the low-frequency vibrations of the lattice. These shifts (the lattice distortions) in turn react, through this non-linear interaction, as a potential well to trap the amide-I vibrational quantum and prevent dispersion of the energy of the amide-I vibrational quantum *via* the dipole-dipole interaction which exists in the neighbouring peptides with certainly electric moments. Thus the vibron-soliton occurs. It is a dynamic self-sustaining entity. The main property of the soliton is it can move over macroscopic distances retaining the wave shape, energy, momentum and other quasi-particles with velocity v. Using this model we have given the red-shift value and other results which are basically consistent with the experimental data [19–21].

There is only the subsonic solitons ($V < V_0, V_0$ is speed of the sound waves of molecular chains) in the system if we take into account only the linear harmonic vibration of the molecular chains [13–19]. However, in the case of the molecular chain, which is in contact with a thermal reservoir at a temperature of $T \neq 0$ K, where the lattice has anharmonic vibrations, a supersonic motion of the soliton appears in the systems [10,16,21]. The supersonic soliton formed can move together with the localized chain deformation along the molecular chains. In molecular crystals where the above mentioned supersonic soliton exists at $T \neq 0$ K, the states of atoms in the molecular chains will be changed due to the interaction between the soliton and the atoms [13,21]. Furthermore, the nuclei situated at crystal sites will also be excited or activated. Thus, gamma-quantum emission of the active nuclei probably appears. This effect will result in an observable Mössbauer effect. In fact, when an active nucleus emits a gamma-ray, the transition energy, ΔE_{nm}, in principle, may be distributed between the gamma-photon and the nucleus that emitted the photon, the chain as a whole, and finally between the vibrations of the chains (in the case of our discussion, instead of pure phonon vibrations, the localized self-trapped solitonic state occurs due to the exciton-phonon interaction). The energy needed for a nucleus to leave its site in the chain is at least 10 eV, but the recoil energy does not exceed several tenths of an electronvolt. As a result, an atom whose nucleus has emitted a gamma-ray cannot change its position in the lattice. The recoil energy which a molecular chain can receive as a whole is small ($N \gg 1$), so that it may be neglected. Thus the transition energy can be distributed only between the gamma-quantum and the solitons. A Mössbauer transition occurs if the solitonic state of the chain is not changed, and the gamma-photon receives the entire energy of the transition. The purpose of this paper is to study the properties of the Mössbauer effect resulting from this mechanism and to calculate corresponding gamma-radiative Mössbauer transition probabilities by means of the vibrational quantum (vibron) model, *i.e.*, we here proposed a new experimental confirmation of the existence of solitons in molecular crystals, which is helpful to understanding the nature of the solitons occurring in the systems and facilitates the development of soliton physics. The plan of this paper is the following. In Section 2 we derive the equations of motion from the given Hamiltonian and wave function [12–21] and give the soliton solutions of these equations in the anharmonically organic molecular crystals at finite temperature $T \neq 0$ K. In Section 3 the gamma-radiative Mössbauer transition probabilities have been obtained and some properties of this effect are also discussed.

2 The soliton excitations in the anharmonical crystals at T $\neq$ 0 K

Due to the fact that we here consider the molecular crystals to be at a finite temperature, which are in contact with a thermal reservoir at temperature of $T \neq 0$ K, there are anharmonic vibrations of the molecular chains in the systems when compared with the case where we do not take into account the temperature effect [16,21]. Therefore, the Hamiltonian describing the collective excitation state resulting from the structural deformation and localized fluctuation in such a case should be represented as [12–21]

$$H = H_{\rm ex} + H_{\rm ph} + H_{\rm int} \tag{1}$$

$$H_{\rm ex} = \frac{1}{2m}\sum_i p_i^2 + \frac{1}{2}m\omega_0^2\sum_i r_i^2 - \frac{1}{2}m\omega_1^2\sum_i r_i r_{i+1} \tag{1a}$$

$$H_{\rm ph} = \frac{1}{2}M\sum_i \dot{R}_i^2 + \sum_i V(R_i - R_{i-1}) \tag{1b}$$

$$H_{\rm int} = \frac{1}{2}m\chi_1\sum_i (R_{i+1} - R_{i-1})r_i^2 + m\chi_2\sum_i (R_{i+1} - R_i)r_i r_{i+1}. \tag{1c}$$

This Hamiltonian represents the properties of the intramolecular excitation caused by the localized fluctuation and the features of sound vibration of the molecular chain arising from the structural deformation and the interaction between the two modes of motion, respectively. Here m and M are the masses of the vibrational quanta (vibron or exciton) generated by the intramolecular excitation and of a molecule in an unit cell, respectively. ω_0 and ω_1 are the diagonal and non-diagonal elements of dynamic matrix, the ω_0 is also the Einstein resonant frequency of the amide-I (C=O stretching) oscillator. $2\chi_1 = \frac{\partial\omega_0^2}{\partial R_i}$ and $2\chi_2 = \frac{\partial\omega_1^2}{\partial R_i}$ are the changes of the vibrational energy of the molecular lattice and of the coupling interaction between neighbouring vibrons by unit extension of the chain. r_i and $p_i = m\dot{r}_i$ are the normal coordinate of the oscillator and its canonical conjugate momentum, respectively. R_i and $P_i = M\dot{R}_i$ are the canonically conjugate operators of displacement and momentum of the molecule, respectively. $V(R_i - R_{i-1})$ is the non-linear interaction potential due to the temperature $T \neq 0$ K, which is usually represented by the Toda lattice potential [21]

$$V(R) = \frac{\alpha'}{\beta}({\rm e}^{-\beta R} - 1) + \alpha' R \approx \frac{1}{2!}\alpha'\beta R^2 - \frac{\alpha'\beta^2}{3!}R^3 + \cdots \tag{2}$$

where $\lambda = \alpha'\beta$ and $\lambda_1 = \frac{\alpha'\beta^2}{2}$ are the elastic and non-elastic constants of the molecular chain, respectively.

Due to the fact that the intramolecular excitation and the vibration of the molecular chain are all quantized then r_i, p_i, R_i and P_i should be represented by corresponding creation and annihilation operators. Thus we here adopt the standard canonical quantized transformations

$$r_i = (2m\omega_0/\hbar)^{-1/2}(b_i^+ + b_i),$$
$$p_i = (\frac{1}{2}\hbar m\omega_0)^{1/2}(-{\rm j})(b_i - b_i^+) \quad ({\rm j} = \sqrt{-1}) \tag{3}$$

$$R_i = \sum_q (\hbar/2MN\omega_q)^{1/2}(a_q + a_{-q}^+){\rm e}^{jir_0 q};$$
$$P_i = {\rm j}\sum_q (M\hbar\omega_q/2N)^{1/2}(a_{-q}^+ - a_q){\rm e}^{jir_0 q} \tag{4}$$

where $\omega_q = 2(\lambda/M)^{1/2}\sin(r_0 q/2)$ is the frequency of phonon with wave vector, q. N is the number of sites in the molecular lattice, r_0 is the distance between the molecules, b_i^+ (b_i) and a_q^+ (a_q) are the creation (annihilation) operators of vibron (exciton) and phonon, respectively. Thus, when $V(R)$ is chosen as a cubic anharmonicity, equation (1) becomes

$$\begin{aligned} H = {} & \sum_i \varepsilon_0(b_i^+ b_i + \frac{1}{2}) - \frac{\hbar\omega_1^2}{4\omega_0}\sum_i (b_i^+ b_{i+1} + b_i b_{i+1}^+) \\ & + \hbar\sum_q \omega_q(a_q^+ a_q + \frac{1}{2}) - \sum_{i,q_1,q_2} F(q_1,q_2)(a_{q_1} + a_{-q_1}^+) \\ & \times (a_{q_2} + a_{-q_2})(a_{(q_1+q_2)} + a_{-(q_1+q_2)}^+) \\ & + \sum_{i,q} [g(q)(b_i^+ b_i + b_i b_i^+) \\ & + g_1(q)(b_i^+ b_{i+1} + b_i b_{i+1}^+)](a_q + a_{-q}^+){\rm e}^{jir_0 q} \end{aligned} \tag{5}$$

where

$$\begin{cases} F(q_1,q_2) = \frac{8}{3}{\rm j}\lambda_1(\frac{\hbar}{2M})^{1/2}(\omega_{q_1}\omega_{q_2}\omega_{(q_1+q_2)})^{-1/2} \\ \qquad \times \sin(\frac{1}{2}r_0 q_1)\sin(\frac{1}{2}r_0 q_2)\sin(\frac{1}{2}r_0(q_1+q_2)); \\ g(q) \quad = (\frac{\hbar}{2MN\omega_q})^{1/2}(\frac{\hbar\chi_1}{4\omega_0})({\rm e}^{{\rm j}r_0 q} - {\rm e}^{-{\rm j}r_0 q}); \\ g_1(q) \quad = (\frac{\hbar}{2MN\omega_q})^{1/2}(\frac{\hbar\chi_2}{2\omega_0})({\rm e}^{{\rm j}r_0 q} - 1); \ \varepsilon_0 = \hbar\omega_0. \end{cases} \tag{6}$$

Due to the fact that the molecular crystal we here study is a closed system which does not exchange particles with the environment, we could ignore these terms including $b_n b_m$ and $b_n^+ b_m^+$ ($m = n, n+1$) because if these terms are retained in the above the Hamiltonian then shows that the number of particles of the system is not conserved which is not appropriate for the system. Therefore we have omitted the above terms in equation (5) when inserting equations (3, 4) in equation (1).

As far as the wave function of the system is concerned, owing to the fact that the non-linear interaction arising from the vibrational quantum (vibron)-phonon interaction can result in not only the coherent excitation of the phonons but also the coherent excitation of the vibrons, namely when the phononic wave function is represented by a coherent state, the vibronic wave function should also be represented by a coherent or quasi-coherent state, instead of a excitation state of a single-particle. After noting this fact and taking into account the thermal distribution of the phonons due to the finite temperature $T \neq 0$ K, thus the wave function describing the states of collective excitations occurring in the organic molecular crystals at a finite temperature should be represented as [12–21]

$$|\Phi_\nu\rangle = |\varphi\rangle|a_\nu\rangle \tag{7}$$

where

$$\begin{aligned}|\varphi\rangle &= \frac{1}{\lambda'}\left(1+\sum_i \varphi_i(t) b_i^+\right)|0\rangle_{\rm ex}\\ &\geq \frac{1}{\lambda'}\exp\left[\sum_i \varphi_i(t) b_i\right]|0\rangle_{\rm ex}\end{aligned} \tag{7a}$$

(we take the normalized factor $\lambda' = 1$ in the following calculation for convenience) and

$$\begin{aligned}|\alpha_\nu\rangle &= U_i^+|\nu\rangle\\ &= \exp\left\{\sum_q [\alpha_{q,i}(t)a_q^+ - \alpha_{q,i}^*(t)a_q]\right\}|\nu\rangle\end{aligned} \tag{7b}$$

while

$$|\nu\rangle = \prod_q (\nu_q'!)^{-1/2}(a_q^+)^{\nu_q'}|0\rangle_{\rm ph} \tag{7c}$$

is the phononic complete set which represents the elementary excitation state of the single phonon due to the finite temperature $T \neq 0$ K. $|0\rangle_{\rm exp}$ and $|0\rangle_{\rm ph}$ are the vacuum states of the vibron and phonon, respectively. We here should point out that equation (7a) is not a standard coherent state and excitation state of single particles, but a coherent superposition of the ground state and one vibronic state, *i.e.*, it is a quasi-coherent state. The reason we adopt it is that the number of particles contained in that state should be determinate (it, in fact, contains only one quantum, *i.e.*, $N = \langle\varphi|(\sum_i b_i^+ b_i)|\varphi\rangle = \sum_i |\varphi_i(t)|^2 = 1$), but a standard coherent state contains innumerable particles which can not maintain the conservation of particles in the system, equation (1) or equation (5). This is not appropriate for the molecular crystal of closed systems. Moreover, owing to the fact that we have already considered the mutual influence between the vibrons and the phonons in this wave function, equation (7) as mentioned above, it again is not necessary to construct the linear correlated wave function of the vibron (exciton) and phonon as the $|D_1\rangle$ state for the Davydov model in the protein molecules [9].

By utilizing the above Hamiltonian, equations (5–7), we can directly derive the equations of motion from the expectation values of the vibron and phonon operators in the state in equation (7): $\varphi_n(t) = \langle\Phi(t)|b_n|\Phi(t)\rangle$ and $\alpha_q(t) = \langle\Phi(t)|a_q|\Phi(t)\rangle$. As a matter of fact, taking again the time derivative of the above variables and assuming that $|\Phi(t)\rangle$ satisfies a nonstationary Schrödinger equation, *i.e.*, $\mathrm{j}\hbar\frac{\partial}{\partial t}|\Phi(t)\rangle = H|\Phi(t)\rangle$, we can thus obtain the required evolution equations

$$\begin{aligned}\mathrm{j}\hbar\frac{\partial}{\partial t}\varphi_n(t) &= \overline{\langle\Phi_\nu|[b_i, H]\Phi_\nu\rangle}\\ \mathrm{j}\hbar\frac{\partial}{\partial t}\alpha_q(t) &= \overline{\langle\Phi_\nu|[a_q, H]\Phi_\nu\rangle}.\end{aligned} \tag{8}$$

These are just the equations of motion for the vibron (exciton) and phonon, respectively. However, because the molecular crystals we study here are in contact with a thermal reservoir at finite temperature of $T \neq 0$ K, after finishing this calculation of the expectation value in quantum mechanics from equation (8), we should calculate the thermal mean value by means of the following formulae:

$$\begin{aligned}\overline{Y} = \langle Y\rangle &= \mathrm{Tr}((\rho_{\nu\nu})_{\rm ph}Y(t))\\ &= \sum_\nu [\langle\nu|\rho|\nu\rangle_{\rm ph}\langle\Phi_\nu|Y(t)|\Phi_\nu\rangle].\end{aligned} \tag{9}$$

The horizontal lines in equations (8, 9) represent the thermal mean value. The density matrix, $\sum_\nu\langle\nu|\rho|\nu\rangle_{\rm ph}$ containing the effect of temperature of $T \neq 0$ K is given in the following form:

$$\begin{aligned}&\langle\nu|\rho|\nu\rangle_{\rm ph} = (\rho_{\nu\nu})_{\rm ph}\\ &\quad= \{\langle\nu|\exp\left[-\frac{H_{\rm ph}}{K_{\rm B}T}\right]|\nu\rangle / \sum_\nu\langle\nu|\exp\left[-\frac{H_{\rm ph}}{K_{\rm B}T}\right]|\nu\rangle\end{aligned} \tag{9a}$$

where the diagonal matrix elements of the Hamiltonian are

$$\langle\Phi_\nu|H|\Phi_\nu\rangle = \langle\Phi_\nu|(H_{\rm ex}+H_{\rm int})|\Phi_\nu\rangle + \langle\alpha_\nu|H_{\rm ph}|\alpha_\nu\rangle. \tag{9b}$$

Inserting equations (5–7) into equation (8), and making use of the following relationship

$$\langle\alpha_\nu|(a_q^+ + a_{-q}^+)|\alpha_\nu\rangle = \alpha_{iq}(t) + \alpha_{i-q}^*(t);$$

$$\langle\alpha_\nu|a_q^+ a_q|\alpha_\nu\rangle = (\nu_q' + |\alpha_{iq}|^2);$$

$$\begin{aligned}&\sum_\nu \exp\left[-\hbar\omega_q\nu_q'/K_{\rm B}T\right]\langle\nu|\exp(\alpha_{iq}^* a_q \exp(-\alpha_{iq}a_q^+)|\nu\rangle\\ &\qquad= (\nu_q+1)\mathrm{e}^{-|\alpha_{iq}|^2(\nu_q+1)};\end{aligned}$$

$$\nu_q = \left[\exp\left(\frac{\hbar\omega_q}{K_{\rm B}T}\right) - 1\right]^{-1};$$

$$\begin{aligned}\exp[-\overline{W}_{i,i\pm1}] &= \langle\alpha_{iq}|\alpha_{(i\pm1)q}\rangle\\ &= \exp\left\{\sum_q[\alpha^*_{(i\pm1)q}\alpha_{iq} - \frac{1}{2}(|\alpha_{(i\pm1)q}|^2 + |\alpha_{iq}|^2)]\right\}\\ &\times\prod_q\left\{\sum_{m=0}^{\nu'_q}\frac{(-1)^m|\alpha_{iq}-\alpha_{(i\pm1)q}|^{2m}\nu'_q!}{(m!)^2(\nu'_q-m)!}\right\},\end{aligned}\tag{10}$$

we get, after some tedious calculations, the equations of motion in the following form:

$$\begin{aligned}\mathrm{j}\hbar\dot{\varphi}_i &= \varepsilon_0\varphi_i - JB(q,T)(\varphi_{i+1}+\varphi_{i-1})\\ &+ \sum_q 2g(q)(\alpha_{iq}+\alpha^*_{iq})\varphi_i\mathrm{e}^{\mathrm{j}ir_0q}\\ &+ \sum_q g_1(q)[(\alpha_{iq}+\alpha^*_{iq})(\varphi_{i+1}+\varphi_{i-1})\mathrm{e}^{\mathrm{j}ir_0q}]\end{aligned}\tag{11}$$

$$\begin{aligned}\mathrm{j}\hbar\dot{\alpha}_{iq} &= \hbar\omega_q\alpha_{iq} + \sum_i 2g(q)|\varphi_i|^2\mathrm{e}^{-\mathrm{j}ir_0q}\\ &+ \sum_q g_1(q)(\varphi^*_i\varphi_{i+1}+\varphi^*_i\varphi_{i-1})\mathrm{e}^{-\mathrm{j}ir_0q}\\ &- \lambda_1\sum_k F(k-q)(\alpha_{ik}+\alpha^*_{i-k})(\alpha^*_{ik-q}+\alpha_{iq-k})\end{aligned}\tag{12}$$

$$\begin{aligned}\mathrm{j}\hbar\dot{\alpha}^*_{i-q} &= -\hbar\omega_q\alpha^*_{i-q} - \Bigg(\sum_i 2g(q)|\varphi_i|^2\mathrm{e}^{-\mathrm{j}ir_0q}\\ &+ \sum_i g_1(q)(\varphi^*_i\varphi_{i+1}+\varphi^*_i\varphi_{i-1})\mathrm{e}^{-\mathrm{j}ir_0q}\\ &-\lambda_1\sum_k F(k-q)(\alpha_{ik}+\alpha^*_{i-k})(\alpha^*_{ik-q}+\alpha_{iq-k})\Bigg)\end{aligned}\tag{13}$$

where

$$\begin{aligned}B(q,T) &\approx \exp[-\overline{W}_i]\\ &\approx \prod_q\exp\left\{-2\coth\left(\frac{\hbar\omega_q}{2K_\mathrm{B}T}\right)|\alpha_{iq}|^2\sin\left(\frac{1}{2}r_0q\right)\right\},\\ J &= \frac{\hbar\omega_1^2}{4\omega_0},\ \varepsilon_0 = \hbar\omega_0.\end{aligned}\tag{14}$$

From equations (12, 13), we obtain

$$\begin{aligned}j(\dot{\alpha}_{iq}+\dot{\alpha}^*_{i-q}) &= -\omega_q(\alpha^*_{i-q}-\alpha_{iq}) \quad \text{or}\\ (\ddot{\alpha}_{i_q}+\ddot{\alpha}^*_{i-q}) &= \mathrm{j}\omega_q(\dot{\alpha}^*_{i-q}-\dot{\alpha}_{iq})\end{aligned}$$

and

$$\begin{aligned}\mathrm{j}\hbar(\dot{\alpha}^*_{i-q}-\dot{\alpha}_{iq}) &= -\hbar\omega_q(\alpha^*_{i-q}+\alpha_{iq})\\ &- \sum_i[4g(g)|\varphi_i|^2 + 2g_1(q)(\varphi^*_i\varphi_{i+1}+\varphi^*_i\varphi_{i-1})]\mathrm{e}^{-\mathrm{j}ir_0q}\\ &+ 2\lambda_1\sum_k(F(k-q)(\alpha_{ik}+\alpha^*_{i-k})(\alpha^*_{ik-q}+\alpha_{iq-k})).\end{aligned}$$

Thus we obtain from the above equations

$$\begin{aligned}(\ddot{\alpha}^*_{i-q}+\ddot{\alpha}_{iq}) &= -\omega_q^2(\alpha^*_{iq}+\alpha_{iq}) - \sum_i\frac{4g(q)\omega_q}{\hbar}|\varphi_i|^2\mathrm{e}^{-\mathrm{j}ir_0q}\\ &- \sum_i\frac{2g_1(q)\omega_q}{\hbar}(\varphi^*_i\varphi_{i+1}+\varphi^*_i\varphi_{i-1})\mathrm{e}^{-\mathrm{j}ir_0q}\\ &+ \frac{2\lambda_1\omega_q}{\hbar}\sum_k F(k-q)(\alpha_{iq}+\alpha^*_{i-k})(\alpha^*_{ik-q}+\alpha_{iq-k}).\end{aligned}\tag{15}$$

Taking into account the relationship between the $u_i(t) = \langle\alpha_\gamma|R_i(t)|\alpha_\gamma\rangle$ and its Fourier variable $u_q(t)$:

$$u_i(t) = N^{-1/2}\sum_q u_{iq}(t)\mathrm{e}^{\mathrm{j}qx} \quad (x = ir_0)\tag{16}$$

where $u_{iq}(t) = \left(\frac{\hbar}{2M\omega_q}\right)^{-1/2}(\alpha_{iq}+\alpha^*_{i-q})$ we have on inserting the above equation into equation (15) the result

$$\begin{aligned}\ddot{u}_{iq}+\omega_q^2u_{iq} &= \frac{\mathrm{j}\hbar\chi_1}{2M\omega_q}\sin(r_0q)|\varphi_i|^2\mathrm{e}^{-\mathrm{j}ir_0q}\\ &+ \frac{\hbar\chi_2}{2M\omega_0N^{1/2}}\sum_i(\mathrm{e}^{\mathrm{j}r_0q}-1)(\varphi^*_i\varphi_{i+1}+\varphi^*_{i-1}\varphi_i)\mathrm{e}^{-\mathrm{j}ir_0q}\\ &- \frac{j8\lambda_1}{3M}\sin\left(\frac{1}{2}r_0q\right)\\ &\times\sum_k\sin\left(\frac{1}{2}r_0k\right)\sin\left[\frac{1}{2}r_0(q-k)\right]u_{ik}u_{iq-k}\mathrm{e}^{-\mathrm{j}(k-q)r_0}.\end{aligned}\tag{17}$$

In the long wave-length limit for low frequency vibrations

$$\begin{gathered}\sin r_0q \sim r_0q,\\ \sin\frac{1}{2}r_0k \sim \frac{1}{2}r_0k,\\ \sin\frac{1}{2}(q-k)r_0 \sim \frac{1}{2}r_0(q-k),\\ (\mathrm{e}^{\mathrm{j}r_0q}-1) \sim \mathrm{j}r_0q\end{gathered}$$

and taking into account the dispersion relation in second order approximation:

$$\begin{aligned}\omega_q &= 2(\lambda/M)^{1/2}\sin\left(\frac{1}{2}r_0q\right)\\ &\approx 2(\lambda/M)^{1/2}\left[\frac{1}{2}r_0q - \frac{1}{3!}\left(\frac{1}{2}r_0q\right)^3\right]\\ &= V_0q\left(1-\frac{1}{24}q^2r_0^2\right)\end{aligned}$$

where $V_0 = (\lambda/M)^{1/2} r_0$ is the speed of sound. Thus equation (17) becomes

$$\begin{aligned}\ddot{u}_{iq} + V_0^2 q^2 u_{iq} - \frac{1}{12} V_0^2 r_0^2 q^4 u_{iq} = &\\ \frac{\mathrm{j}\hbar\chi_1}{2M\omega_0 N^{1/2}}(r_0 q)\sum_i |\varphi_i|^2 \mathrm{e}^{-\mathrm{j}ir_0 q} &\\ + \frac{\hbar\chi_2}{2M\omega_0 N^{1/2}}\sum_i (r_0 q)(\varphi_i^* \varphi_{i+1} + \varphi_i^* \varphi_{i-1})\mathrm{e}^{-\mathrm{j}ir_0 q} &\\ - \frac{j\lambda_1 r_0^3 q}{3M}\sum_k (q-k) k u_{ik} u_{iq-k}. &\qquad (18)\end{aligned}$$

Multiplying equation (18) with $N^{-\frac{1}{2}} \mathrm{e}^{\mathrm{j}r_0 iq}$ and summing over the wave number, q, at the same time, and making the continuum approximation:

$$\varphi_{i\pm1} = \varphi_i \pm r_0 \frac{\partial}{\partial x}\varphi_i + \frac{1}{2!} r_0^2 \frac{\partial^2}{\partial x^2}\varphi_i + \ldots, \qquad \varphi_i(t) \to \varphi(x,t)$$

$$u_{i\pm1} = u_i \pm r_0 \frac{\partial}{\partial x} u_i + \frac{1}{2!} r_0^2 \frac{\partial^2}{\partial x^2} u_i + \cdots, \qquad u_i(t) \to u(x,t)$$

and again utilizing equation (16) and

$$\frac{\partial}{\partial x} u(x,t) = N^{1/2} \sum_q (\mathrm{j}q) u_{iq}(t) \mathrm{e}^{\mathrm{j}qx}$$

and

$$\frac{\partial^2}{\partial x^2} u(x,t) = N^{1/2} \sum_q (\mathrm{j}q)^2 u_{iq}(t) \mathrm{e}^{\mathrm{j}qx}, \text{ etc.}$$

We finally obtain

$$\begin{aligned}\frac{\partial^2 u}{\partial t^2} - V_0^2 \frac{\partial^2 u}{\partial x^2} - \frac{v_0^2 r_0^2}{12}\frac{\partial^4 u}{\partial x^4} = &\\ \frac{\hbar r_0(\chi_1+\chi_2)}{2M\omega_0}\frac{\partial}{\partial x}|\varphi|^2 + \frac{\lambda_1 r_0^3}{3M}\frac{\partial}{\partial x}\left|\frac{\partial}{\partial x} u\right|^2 &\qquad (19)\end{aligned}$$

$$\begin{aligned}\mathrm{j}\hbar\frac{\partial}{\partial t}\varphi(x,t) = (\varepsilon_0 - 2JB(q,T))\varphi(x,t) &\\ - Jr_0^2 B(q,T)\frac{\partial^2}{\partial x^2}\varphi(x,t) &\\ + \frac{\hbar(\chi_1+\chi_2)r_0}{\omega_0}\frac{\partial u(x,t)}{\partial x}\varphi(x,t). &\qquad (20)\end{aligned}$$

Equations (19, 20) are a complete set of equations of motion for the vibron and the phonon generated in the organic molecular crystal at temperatures $T \neq 0$ K in the continuum approximation. Because the $B(T,q)$ relates to temperatures $T \neq 0$ K, the properties of the soliton which are determined by equations (19, 20) depend critically on the temperature. However, finding its soliton solutions is very difficult. We can find the analytic solutions by using approximation and iteration.

2.1 The approximate method

Let us assume the solution of equation (19) to be of the form

$$Q(x,t) = Q_0(x,t) + Q_1(x,t)$$

where $Q(x,t) = -\frac{\partial u}{\partial x}$, and $Q_0(x,t) = -\frac{\partial u_0}{\partial x}$ satisfies the following equation:

$$\ddot{Q}_0(x,t) - V^2\left(\frac{\partial^2 Q_0(x,t)}{\partial x^2} + \frac{r_0^2}{12}\frac{\partial^4 Q_0(x,t)}{\partial x^4}\right) = \frac{\lambda_1 r_0^3}{3M}\frac{\partial^2}{\partial x^2}|Q_0(x,t)|^2.$$

Setting $\xi = x - Vt$, we obtain from the previous equation

$$(V^2 - V_0^2)\frac{\partial^2 Q_0}{\partial \zeta^2} - \frac{r_0^2 V_0^2}{12}\frac{\partial^4 Q_0}{\partial \zeta^4} = \frac{\lambda_1 r_0^2}{3M}\frac{\partial^2}{\partial \zeta^2} Q_0^2\,.$$

After making two integrations and having in mind boundary conditions

$$\varphi(\pm\infty) = \varphi_x(\pm\infty) = u(\pm\infty) = Q(\pm\infty) = 0 \qquad (21)$$

we get

$$\frac{1}{12} r_0^2 V_0^2 \frac{\partial^2 Q_0}{\partial \zeta^2} - (V^2 - V_0^2) Q_0 + \frac{\lambda_1 r_0^3}{3M} Q_0^2 = 0.$$

After again twice integrating the above equation, we obtain the solution [12,15,16]:

$$\begin{aligned}Q_0(\zeta) &= Q_0(x,t)\\ &= \frac{3M(V^2 - V_0^2)}{4\lambda_1 r_0^3}\mathrm{sech}^2\left[\frac{1}{r_0}\left(\frac{V^2}{2V_0^2} - \frac{1}{2}\right)^{-1/2}(x - Vt)\right],\\ &\qquad (V > V_0). \qquad (22)\end{aligned}$$

Now substituting equation (22) into equation (20), we obtain

$$\mathrm{j}\hbar\frac{\partial\varphi}{\partial t} = E_0\varphi - \frac{\hbar^2}{2m^*(T)}\frac{\partial^2\varphi}{\partial x^2} - W_0 \mathrm{sech}^2(\nu\zeta)\varphi(\zeta) \qquad (23)$$

where

$$\begin{aligned}E_0 &= [\varepsilon_0 - 2JB(q,T)],\\ \frac{\hbar^2}{2m^*(T)} &= Jr_0^2 B(q,T);\\ \nu &= \frac{1}{r_0}\left(\frac{V^2}{2V_0^2} - \frac{1}{2}\right)^{-1/2},\\ W_0 &= \frac{3\hbar M(V^2 - V_0^2)(\chi_1+\chi_2)}{4\lambda_1\omega_0 r_0^2},\\ m^*(T) &= mB(q,T),\\ S &= V/V_0.\end{aligned}$$

Now we assume that the solution of equation (23) is of the form [19,23]

$$\varphi(x,t)=\phi(\zeta)\exp[\mathrm{j}(\overline{k}x-\omega_{\mathrm{sol}}t)] \tag{24}$$

where the solitonic wave-vector $\overline{k}=m^*V/\hbar$ and the solitonic energy $E_{\mathrm{sol}}=\hbar\omega_{\mathrm{sol}}$. Inserting equation (24) into equation (23), the equation satisfied by the real part of the amplitude, $\phi(\zeta)$, is

$$\frac{\mathrm{d}^2\phi(\zeta)}{\mathrm{d}\zeta^2}-\frac{2m^*(T)}{\hbar^2}(E-W_0\mathrm{sech}^2(\nu\zeta))\phi(\zeta)=0 \tag{25}$$

where $E=-\hbar\omega_{\mathrm{sol}}+E_0+\frac{1}{2}m^*V^2$. When $\hbar^2\nu^2/m^*=W_0=2E$, the solution of equation (25) is [21–23]

$$\phi(\zeta)=[W_0m^*(T)/2\hbar^2]^{1/2}\mathrm{sech}(\nu\zeta).$$

Thus

$$\begin{aligned}\varphi(x,t)&=\left(\frac{W_0m^*(T)}{2\hbar^2}\right)^{1/2}\mathrm{sech}[\nu(x-Vt)]\mathrm{e}^{\mathrm{j}(\overline{k}x-\omega_{\mathrm{sol}}t)}\\&=A(T)\mathrm{sech}[\nu(x-Vt)]\mathrm{e}^{\mathrm{j}(\overline{k}x-\omega_{\mathrm{sol}}t)}.\end{aligned} \tag{26}$$

In the general case, the solution of equation (25) is $P_{g'}^{\mu'}(\tanh(\nu\zeta))$ where $P_{g'}^{\mu'}$ is the associated Legendre function. In the special case where $\mu'=g'$, the solution reduces to

$$\varphi(x,t)=A'(T)\mathrm{sech}^{\mathrm{g}'}[\nu(x-Vt)]\mathrm{e}^{\mathrm{j}(\overline{k}-\omega_{\mathrm{sol}}t)} \tag{27}$$

where $A'(T)$ is a constant of normalization which depends indirectly on the temperature as

$$A'(T)=\nu\Gamma\left[g'(T)+\frac{1}{2}\right]\Big/\sqrt{\pi}\Gamma[g'(T)]$$

where $\Gamma(x)$ is the gamma function, the parameter $g'(T)$ is an integer which depends on the temperature T and it satisfies the condition:

$$g'(T)=g'\le\frac{1}{2}\left\{\left[1+\frac{12r_0^2\lambda(\chi_1+\chi_2)}{\hbar\lambda_1\omega_0}m^*(T)\right]-1\right\}$$

$$\text{and } \mu'=\left(\frac{2m^*(T)E}{\hbar^2\nu^2}\right)^{1/2}.$$

Equations (26, 27) are just a supersonic soliton with $s>1$ $(V>V_0)$. Its speed and amplitude depend on the anharmonicity λ and temperature T through $m^*(T)$ and $A(T)$. To find an explicit expression for $\varphi(x,t)$, we must determine the representation of $B(T,q)$.

From equation (4), we can obtain

$$a_{iq}=\frac{1}{\mathrm{j}r_0q}\left(\frac{M\omega_q}{2\hbar}\right)^{1/2}(Q_{iq}-\dot{Q}_{iq}/\mathrm{j}\omega_q). \tag{28}$$

We use here the method of the modulated waves, *i.e.*,

$$\begin{aligned}a_{iq}&=a_{iq}(t)\mathrm{e}^{-\mathrm{j}xq},\\Q_{iq}&=Q_{iq}(t)\mathrm{e}^{-\mathrm{j}xq},\\Q_i(t)&=N^{-1/2}\sum_q Q_{iq}(t)\mathrm{e}^{-\mathrm{j}xq},\quad (x=ir_0).\end{aligned}$$

Utilizing equation (16) and $Q(x,t)=-\frac{\partial u(x,t)}{\partial x}$, we may get

$$Q_{iq}(t)=Q_i(q,t)=N^{-1/2}\int_{-\infty}^{\infty}\frac{\mathrm{d}x}{r_0}Q(x,t)\mathrm{e}^{-\mathrm{j}qx}.$$

Inserting equation (23) into the above equation, and again integrating, we obtain

$$\begin{aligned}Q_{iq}&=Q_i(q,t)\\&=N^{-1/2}\frac{3M(V^2-V_0^2)}{4\lambda_1r_0^2}\int_{-\infty}^{\infty}\mathrm{sech}^2(\nu(x-Vt))\mathrm{e}^{-\mathrm{j}qx}\mathrm{d}x\\&=\frac{N^{-1/2}3\pi MV_0^2q}{2\lambda_1r_0}\mathrm{sech}\left(\frac{\pi qr_0}{2\nu}\right)\mathrm{e}^{-jqVt}.\end{aligned}$$

Thus we can get

$$|a_{iq}|^2=(9\pi^2M^3V_0^3q(V^2+V_0^2)/8N\lambda_1^2r_0\hbar)\mathrm{sech}^2(\pi q/2\nu).$$

Substituting the above equation into equation (14), after some calculation as above, we finally obtain

$$\begin{aligned}B(q,T)=\prod_q\exp\Bigg\{&-\frac{9\pi^2M^3V_0^3q(V^2+V_0^2)}{4\hbar N\lambda_1^2r_0^4}\\&\times\mathrm{cth}\left(\frac{\hbar\omega_q}{2K_{\mathrm{B}}T}\right)\mathrm{sech}^2\left(\frac{\pi qr_0}{2\nu}\right)\sin^2\left(\frac{1}{2}r_0q\right)\Bigg\}\end{aligned} \tag{29a}$$

or

$$\begin{aligned}B(q,T)=\exp\Bigg\{-\Bigg[&\frac{9M^3V_0^3(V_0^2+V^2)K_{\mathrm{B}}T\nu^2}{4\hbar^2r_0^3N\lambda_1^2}\\&\times\sum_{l=0}^{\infty}\frac{2^{2l}B_{2l}((\hbar V_0/2K_{\mathrm{B}}T)q)^{2l}}{(2\ell+1)(2l)!}\Bigg]\Bigg\}\end{aligned} \tag{29b}$$

where B_{2l} is the Bernoulli number.

2.2 The iteration method

Now we assume that the solution of equation (16) is of the form [21–23]

$$\varphi(x,t)=\phi(x,t)\mathrm{e}^{\left(\frac{\mathrm{j}\hbar V}{2J'}(x-x_0)-\mathrm{j}Et/\hbar\right)},\quad (J'=Jr_0^2). \tag{30}$$

Substituting equation (30) into equation (20), we get

$$\begin{aligned}&-Jr_0^2B(q,T)\frac{\partial^2\phi}{\partial x^2}-\frac{\hbar(\chi_1+\chi_2)r_0}{\omega_0}Q\phi=\\&\qquad\left[E-(\varepsilon_0-2JB(q,T)-\frac{\hbar^2V^2B(q,T)}{AJr_0^2}\right]\phi\end{aligned}$$

or

$$-\frac{\partial^2\phi}{\partial x^2}-\rho Q\phi=\varepsilon\phi \tag{31}$$

where $Q = -\frac{\partial u}{\partial x}$, letting $\zeta = x - Vt$, equation (19) becomes

$$(V_0^2 - V^2)\frac{\partial^2 Q}{\partial \zeta^2} + \frac{V_0^2 r_0^2}{12}\frac{\partial^4 Q}{\partial \zeta^4} = \frac{\hbar r_0(\chi_1+\chi_2)}{M\omega_0}\frac{\partial^2}{\partial \zeta^2}|\phi|^2 - \frac{\lambda_1 r_0^3}{3M}\frac{\partial^2 Q^2}{\partial \zeta^2}\cdot$$

Using the double integration for the above equation and utilizing equation (21), we obtain

$$\overline{\nu}\frac{\mathrm{d}^2 Q}{\mathrm{d}\zeta^2} + (1-s^2)Q + \alpha|Q|^2 = \sigma|\phi|^2. \quad (32)$$

The $\phi(x,t)$ satisfies a normalized condition:

$$\int |\phi|^2 \mathrm{d}\zeta = 1 \quad (33)$$

where

$$\rho = \frac{\hbar(\chi_1+\chi_2)}{J\omega_0 r_0 B(q,T)}, \quad \overline{\nu} = \frac{r_0}{12},$$
$$\varepsilon = \frac{1}{J r_0^2 B(q,T)}\Big[E - (\varepsilon_0 - 2JB(q,T)) - \frac{\hbar^2 V^2 B(q,T)}{4J r_0^2}\Big],$$
$$\sigma = \frac{\hbar r_0(\chi_1+\chi_2)}{2M\omega_0 V^2},$$
$$\alpha = \frac{\lambda_1 r_0^3}{3MV_0^2}, \quad s = V/V_0.$$

Now multiplying equation (31) by $\frac{\partial \phi}{\partial x}$ and equation (32) by $\frac{\partial \phi}{\partial x}$, and after, again integrating the two equations using the equation (21), we get

$$\overline{\nu}\left(\frac{\mathrm{d}Q}{\mathrm{d}\zeta}\right)^2 + (1-s^2)Q^2 + \frac{2}{3}\alpha Q^3 - 2\sigma\phi^2 Q + 4\sigma\int_0^\phi Q(Y)Y\mathrm{d}Y = 0 \quad (34)$$

$$\left(\frac{\mathrm{d}\phi}{\mathrm{d}\zeta}\right)^2 + 2\rho\int_0^\phi Q(Y)Y\mathrm{d}Y + \varepsilon\phi^2 = 0. \quad (35)$$

Obviously, for bell-shaped soliton solutions centered at $\zeta = 0$ there certainly is $\left(\frac{\mathrm{d}\phi}{\mathrm{d}\zeta}\right)|_{\zeta=0} = 0$ [12–21]. Thus we may immediately get from equation (35) the value of the eigenenergy parameter

$$\varepsilon = -2\rho\phi_0^{-2}\int_0^{\phi_0} Q(Y)Y\mathrm{d}Y = -2\rho\int_0^1 Q(y)y\mathrm{d}y, \quad (y = Y/\phi_0) \quad (36)$$

which corresponds to the value of the bound state of the vibron. Inserting this value into equation (35), we may obtain

$$\zeta = \pm(2\rho)^{-1/2}\int_\phi^{\phi_0}\{(\tau^2-\phi_0^2)\int_{\tau/\phi_0}^1 Q(y)y\mathrm{d}y\}^{-1/2}\mathrm{d}\tau. \quad (37)$$

Again using equation (37), we obtain from equation (34)

$$Q(\phi) = \int_0^\phi \left\{\frac{(s^2-1)Q - \frac{2}{3}\alpha Q^3 + 2\sigma\phi^2 Q - 4\alpha\int_0^\tau Q(z)z\mathrm{d}z}{2\rho\overline{\nu}(\phi^2-\phi_0^2)\int_{\tau/\phi_0}^1 Q(z)z\mathrm{d}z}\right\}^{1/2}\mathrm{d}\tau. \quad (38)$$

The unknown amplitude ϕ is determined by the condition of equation (33) which can also be represented as the following form through inserting equation (37) into equation (33)

$$(2\rho)^{-1/2}\int_0^\phi y^2\left\{(y^2-\phi_0^2)\int_{\phi/\phi_0}^{\phi/\phi_0} Q(z)z\mathrm{d}z\right\}^{1/2}\mathrm{d}y = \frac{1}{2}\cdot \quad (39)$$

Thereupon, we can easily get from equation (37) the envelope function $\phi(\zeta) = \phi(-\zeta)$ in the implicit form by having found a solution ϕ_0 and $Q(\phi_0)$ of equations (34, 35). Therefore equations (37–39) are just a set of formulae for finding the solutions of equations (34, 35). Now we are going to solve the integral equations (37–39) by using iteration.

To solve the integral equation (38) by iteration we first choose the function

$$Q^{(0)} = D\phi^2 \quad (40)$$

where D is some positive numbers to be determined below. Inserting equation (40) into equation (39) we get the corresponding value of the amplitude to be in the form of

$$\phi^{(0)} = \frac{1}{2}(D\rho/2)^{1/2}. \quad (41)$$

Inserting again equations (40, 41) into the right-hand side of equation (38) we can get

$$Q^{(1)}(\phi) = \left(\frac{2}{\overline{\nu}\rho}\right)^{1/2}\int_0^\phi\left(\frac{\sigma + D(s^2-1) - \frac{2}{3}D^2\alpha x^2}{\left(\frac{1}{8}D\rho - x^2\right)}\right)x\mathrm{d}x. \quad (42)$$

Obviously, the value of $Q^{(1)}(\phi)$ depends on practical form of D. We now choose D to be a positive root of the following equation

$$\alpha\rho D^3 + 12(1-s^2)D - 12\sigma = 0. \quad (43)$$

Utilizing the constraint condition of D, equation (43), we get from the integral equation (42)

$$Q^{(1)}(\phi) = Dl\phi^2 = lQ^{(0)}(\phi) \quad (44)$$

where $l = \left(\frac{a}{\overline{\nu}\rho} - \frac{\sigma}{\overline{\nu}\rho}D^{-2}\right)^{1/2}$. Inserting equation (44) into equation (37), yields

$$\phi^{(1)} = (Dl\rho/8)^{1/2} = l^{1/2}\phi^{(0)}. \tag{45}$$

Similarly, inserting the first approximation given by equations (44, 45) into the right-hand side of equation (38) and utilizing then equation (39) we can find the second approximation. The higher approximations can be obtained in the same way. It is easy to see that for $l = 1$ the initial function equation (40) with D satisfying equation (39) is a fixed point of the integral equation (38). So if $l = 1$, the expressions (44, 45) with D, a positive root of the equation (43), describe the exact soliton solution with the bell-shaped profiles $\phi(\zeta)$ and $Q(\zeta)$, Near $l = 1$, equations (44, 45) also represent the approximate soliton solution.

It should be noted that the first approximation of the soliton solution given by equations (44, 45) can be written in an explicit form. Indeed, inserting equations (44, 45) into equations (36, 39), we obtain

$$\varepsilon = -(D\ell\rho/4)^2 = -(D\rho/4)^{1/2} = -C(T) \tag{46}$$

$$\begin{aligned}\phi^{(1)}(\zeta) &= (D\rho/8)^{1/2}\mathrm{sech}(D\rho\zeta/4)\\ &= (C(T)/2)^{1/2}\mathrm{sech}\ (C(T)(x - Vt))\\ &= \phi(\zeta) = \phi(x,t)\end{aligned} \tag{47}$$

and

$$\begin{aligned}Q^{(1)}(\zeta) &= \frac{1}{2}C^2(T)\mathrm{sech}^2(C(T)(x - Vt))\\ &= Q(\zeta) = Q(x,t).\end{aligned} \tag{48}$$

From equation (43) we find the positive root of equation (43) to be

$$D = \left(\frac{6\sigma}{\alpha\rho}\right)^{1/3}\left[\left[1 + \left(1 - \frac{16(s^2-1)\overline{\nu}}{3\alpha^2\sigma^2}\right)^{1/2}\right]^{1/3} + \left[1 - \left(\frac{16(s^2-1)\overline{\nu}}{3\alpha^2\sigma^2}\right)^{1/2}\right]^{1/3}\right] > 0$$

therefore, when $3\alpha^2\sigma^2 \geq 16(s^2 - 1)\overline{\nu} > 0$, there is a supersonic soliton with $s > 1$ ($V > V_0$) in equation (47). In such a case, the non-linear vibration of molecular chains exists certainly in the molecular crystal where the above mentioned supersonic solitonic solutions exist at $T \neq 0$ K (*i.e.*, $\alpha > 0$), simultaneously. Thus, we can conclude from equations (47, 27, 28) that the anharmonicity is a necessary condition for forming the supersonic soliton; in other words, if only the supersonic soliton excitation can occur in such a system, then the anharmonic vibration of molecular chains certainly exists. The amplitude of the soliton formed in the molecular crystal in such a case increases as the temperature increases in a complicated fashion and decreases as the anharmonicity coefficient increases in the form of $\alpha^{-1/n}$ ($n \geq 6$). However, if we let J and $(\chi_1 + \chi_2)$ to tend to zero in such a way that $(\chi_1 + \chi_2)^2 J^{-1} \to 0$, while the constraint $\alpha = 3\overline{\nu}\rho$ (*i.e.*, $l = 1$) remains valid, in this limit we find from equation (43) that $D = \frac{2}{\rho}\left(\frac{s^2-1}{\overline{\nu}}\right)^{1/2} > 0, (s > 1)$. Thus the supersonic soliton is of the form

$$\begin{aligned}\phi(\zeta) &= \frac{1}{2}\left(\frac{s^2-1}{\overline{\nu}}\right)^{1/4}\mathrm{sech}\left(\left(\frac{s^2-1}{4\overline{\nu}}\right)^{1/2}\zeta\right)\\ &= \frac{1}{2}\left(\frac{s^2-1}{\overline{\nu}}\right)^{1/4}\mathrm{sech}\left(\left(\frac{s^2-1}{4\overline{\nu}}\right)^{1/2}(x - Vt)\right)\end{aligned} \tag{49a}$$

$$\begin{aligned}Q(\zeta) &= \frac{3(s^2-1)}{2\alpha}\mathrm{sech}^2\left(\left(\frac{s^2-1}{4\overline{\nu}}\right)^{1/2}\zeta\right)\\ &= \frac{3(s^2-1)}{2\alpha}\mathrm{sech}^2\left(\left(\frac{s^2-1}{4\overline{\nu}}\right)^{1/2}(x - Vt)\right)\end{aligned} \tag{49b}$$

where $C = \frac{1}{2}\left(\frac{s^2-1}{\overline{\nu}}\right)^{1/2}$. The formation of these solitons in such a case is due to the balancing of effects between dispersion ν and anharmonicity α, but the amplitude of the soliton depends only on the velocity V in the form of $s^{1/2}$.

When $s = 1$ there is a positive root. In such a case, we have $D = \left(\frac{12\sigma}{\alpha\rho}\right)^{1/3} > 0$. Thus equations (47, 48) become

$$\begin{cases}\phi(\zeta) = \left(\dfrac{3}{124}\right)^{1/6}\left(\dfrac{\sigma\rho^2}{\alpha}\right)^{1/6}\mathrm{sech}\left(\left(\dfrac{3\sigma\rho}{16\alpha}\right)^{1/3}\zeta\right)\\ Q(\zeta) = \left(\dfrac{9\sigma^2\rho}{32\alpha^2}\right)^{1/3}\mathrm{sech}^2\left(\left(\dfrac{3\sigma\rho}{16\alpha}\right)^{1/3}\zeta\right)\end{cases}. \tag{50}$$

This is a sonic soliton. Obviously, its amplitude decreases as the anharmonicity increases in the form $\alpha^{-1/6}$ or $\lambda^{-1/6}$. Also, from the results just obtained, *i.e.*, equations (27, 28, 47, 44), we can see that the influence of the anharmonicity on the amplitude of the soliton is contrary to the temperature $T \neq 0$ K, namely, the anharmonicity makes its amplitude decrease, but the temperature make its amplitude increase. So, we may suppose that the soliton excited in the molecular crystal with temperature $T \neq 0$ K is stable. $B(q,T)$ may be calculated by analogy with the method mentioned above in this case, and the result of $B(q,T)$ is also analogous to equation (29) and is not here displayed.

3 Calculation for the gamma-radiative Mössbauer transition probability

From the above results we see that the supersonic soliton is evolved also from a self-trapped exciton through interaction with an anharmonic vibration of molecular chains in the crystals at temperature $T \neq 0$ K. When the supersonic soliton moves and propagates at the velocity V, which is greater than the sound velocity V_0, along the molecular chains, the states and positions of atoms will be changed, at the same time. Then the activation of nuclei will also appear due to the interaction between the soliton and the atoms. Thus, the active nuclei may emit gamma-photons and the Mössbauer effect will occur as described in the introduction. We now calculate the gamma-radiative Mössbauer transition probability from the active nuclei located in molecular chain node i.

According to quantum theory the interaction potential caused by the emission mentioned above may be represented as [24–27]

$$V = A\Pi(x_\mathrm{L}, P_\mathrm{L}, \sigma_\mathrm{L})\Gamma(P, u_{i_0}) \tag{51}$$

where $\Pi(x_\mathrm{L}, p_\mathrm{L}, \sigma_\mathrm{L})$ is an operator related to the inner state, coordinate x_L, momentum p_L and spin σ_L, of the active nucleus. $\Gamma(P, u_{i_0})$ is an operator related to the emitting state of the active nucleus located at the node i_0 while P is momentum corresponding to the emitted gamma-quantum. Obviously, there exists the relationship:

$$u_{i_0} = i_0 r_0 + R_{i_0} \tag{52}$$

in the emitted process, where R_{i_0} denotes the recoil displacement of the emitting nucleus from its node. In the general case, the $\Gamma(P, u_{i_0})$ may be represented by a periodic potential or a plan wave [24–27], *i.e.*, $\Gamma(P, u_{i_0}) = d\exp(\mathrm{j}P, u_{i_0}/h)$. Thus equation (51) becomes

$$V_\mathrm{int} = \overline{A}\Pi(x_\mathrm{L}, P_\mathrm{L}, \sigma_\mathrm{L})\exp(\mathrm{j}\mathbf{P}\cdot i_0\mathbf{r}_0/\hbar)\exp(\mathrm{j}\mathbf{P}\cdot\mathbf{R}_{i_0}/\hbar) \tag{53}$$

where d and A' are proportionality coefficients which relate the characteristics of the molecular chain and nucleus. In particular, let $\overline{A} = \mathrm{d}A'$. Equation (53) shows that when the emission of gamma-quantum with momentum P occurs the state of the molecular chain will also be changed. According to the quantum theory of radiation [17], the transitive matrix element for the change of state may be represented as

$$\begin{aligned} T_{n\to m} &= \langle m\Phi_\nu^m|V_\mathrm{int}|n\Phi_\nu^n\rangle \\ &= \overline{A}\langle\Phi_\nu^m|\exp(\mathrm{j}\mathbf{P}\cdot\mathbf{R}_{i_0})\exp(\mathrm{j}i_0\mathbf{r}_0\cdot\mathbf{P}/\hbar)|\Phi_\nu^m\rangle \\ &\quad\times\langle m|\Pi(x_\mathrm{L}, P_\mathrm{L}, \sigma_\mathrm{L})|n\rangle. \end{aligned} \tag{54}$$

Keeping in mind that the $\Pi(x_\mathrm{L}, P_\mathrm{L}, \sigma_\mathrm{L})$ depends only on the fixed numbers of internal degrees of freedom of nucleus, it is not necessary to know its explicit form, when we discuss only the emission phenomenon of the organic molecular crystal. Thus the matrix element, $\langle m|\Pi(x_\mathrm{L}, P_\mathrm{L}, \sigma_\mathrm{L})|n\rangle$, in equation (54) may be treated as having a constant value, when the change of the inner state of the nucleus is small, and we let it be absorbed into the constant $\overline{A}$. Due to the reasons mentioned above we are interested only in relative transition probability determined by the following matrix element

$$T_{n\to m} = \overline{A}\langle\Phi_\nu|\exp(\mathrm{j}\mathbf{P}\cdot\mathbf{R}_{i_0}/\hbar)|\Phi_\nu\rangle\exp(\mathrm{j}\mathbf{P}\cdot i_0\mathbf{r}_0/\hbar) \tag{55}$$

i.e., by the transition amplitude in our case when the same soliton is found in the molecular crystal before and after the emission. In other words, the state of the supersonic soliton is not affected by the emission as mentioned in the introduction. Remembering that the $|\Phi\rangle$ represents the soliton amplitude, the R_{i_0} is given by

$$R_{i_0} = \sum_q(\hbar/2NM\omega_q)^{1/2}\mathbf{e}_q(a_q + a_{-q}^+)\exp(\mathrm{j}i_0r_0q) \tag{55a}$$

here the $\mathbf{e}_q$ is the polarization vector of longitudinal phonon.

Inserting equation (7) into equation (55) we obtain

$$T_{n\to m} = \overline{A}\sum_i(1 + |\varphi_i|^2)T_{\nu\nu}(i)\mathrm{e}^{\mathrm{j}i_0r_0P/\hbar}$$

where

$$\begin{aligned} T_{\nu\nu}(i) = \langle a_\nu|\exp\Bigg\{\frac{\mathrm{j}}{\hbar}\sum_q\left(\frac{\hbar}{2MN\omega_q}\right)^{1/2} \\ \times(\mathbf{P}\cdot\mathbf{e}_q)(a_q + a_{-q}^+)\mathrm{e}^{\mathrm{j}i_0r_0q}\Bigg\}|a_\nu\rangle. \end{aligned}$$

Inserting equation (7c) into the above formula and performing the corresponding calculation,we get

$$\begin{aligned} T_{\nu\nu}(i) = \prod_q\exp\left\{\beta_q^*\alpha_{iq}^* - \beta_q\alpha_{iq} + |\beta_q|^2/2\right\} \\ \times\sum_{i_q=0}^{\infty}\frac{(-1)^{i_q}|\beta_q|^{2i_q}(i_q + \nu_q)!}{(i_q!)^2\nu_q!} \end{aligned} \tag{56}$$

where

$$\beta_q = \frac{\mathrm{j}}{\hbar}(\hbar/2MN\omega_q)^{1/2}(\mathbf{P}\cdot\mathbf{e}_q)\mathrm{e}^{\mathrm{j}i_0r_0q}\text{ and }\beta_{-q} = -\beta_q^*.$$

Due to the fact that the molecular crystal we here study is in contact with a thermal reservoir at temperature $T \neq 0$ K, we should take the thermal average over the phonon states for the matrix element equation (55) by using the density matrix equation (9a), *i.e.*,

$$\begin{aligned} \overline{\langle T_{n\to m}\rangle} &= \overline{\overline{A}\sum_i(1 + |\varphi_i|^2)T_{\nu\nu}(i)\mathrm{e}^{\mathrm{j}i_0r_0p/h}} \\ &= \overline{A}\sum_i(1 + |\varphi_i|^2)\langle T_{\nu\nu}(i)\rangle_\mathrm{ph}\mathrm{e}^{\mathrm{j}i_0r_0p/h} \end{aligned} \tag{57}$$

where

$$\begin{aligned} \langle T_{\nu\nu}(i)\rangle_\mathrm{ph} &= \sum_\nu\rho_{\nu\nu}T_{\nu\nu}(i) \\ &= \langle T_\mathrm{sol}\rangle_\mathrm{ph}\exp\left\{-\sum_q\frac{(\mathbf{P}\cdot\mathbf{e}_q)^2}{2MN\omega_q}\left(\nu_q + \frac{1}{2}\right)\right\} \end{aligned} \tag{58}$$

and a_{iq} and a_{iq}^* in equation (56) should be obtained from equations (13–22), or equations (22, 28). Obviously, it is related to the supersonic solitonic solution $\varphi(t)$. Therefore, $\langle T_{\rm sol}\rangle_{\rm ph}$ contains the matrix element for the solitonic part, Inserting the a_{iq}, a^*_{iq} and $\varphi_i(t)$ obtained from previous equations into equation (58) and making the calculation, then the solitonic part of the matrix element, $\langle T_{\rm sol}\rangle_{\rm ph}$, is expressed as

$$\langle T_{\rm sol}\rangle_{\rm ph} = \exp\left\{-\frac{1}{N\hbar}\sum_q \frac{3MNV_0^2}{2\lambda_1 r_0^2}(\mathbf{p}\cdot\mathbf{e}_q) \times\frac{1}{{\rm sh}(\pi q r_0/2\nu)}\exp[-{\rm j}(i_0-i)r_0 q - {\rm j}qVt]\right\}.$$

Due to the fact that in the case of longitudinal phonons the polarization vector is an even function of wavevector ($\mathbf{e}_q = \mathbf{e}_{-q}$) and that the sum of

$$(\mathbf{P}\cdot\mathbf{e}_q)\cos(((i_0-i)r_0+Vt)q)/{\rm sh}(\pi q r_0/2\nu)$$

over the wave-vector q should be zero because it is an odd function of q, then we find

$$\langle T_{\rm sol}\rangle_{\rm ph} = \exp\left\{-\frac{\rm j}{N\hbar}\sum_q \frac{3MNV_0^2}{2\lambda_1 r_0^2}(\mathbf{p}\cdot\mathbf{e}_q) \times\frac{1}{{\rm sh}(\pi q r_0/2\nu)}\sin((i-i_0)r_0 q + Vqt)\right\} \quad (59)$$

where we have adopted the Weyl formula in above calculation, *i.e.*,

$$\begin{aligned} &{\rm e}^{\hat{A}}{\rm e}^{\hat{B}} = {\rm e}^{-\frac{1}{2}[\hat{A},\hat{B}]}{\rm e}^{\hat{A}+\hat{B}} \\ \text{if}\quad &[\hat{A},[\hat{A},\hat{B}]] = [\hat{B},[\hat{B},\hat{A}]] = 0. \end{aligned}$$

The origin of coordinates is now chosen in the active nucleus ($i_0 = 0$) and the sum over q replaced by the integration

$$\frac{1}{N}\sum_q \to \frac{r_0}{2\pi}\int_{-\pi/r_0}^{\pi/r_0}{\rm d}q.$$

In the next step we consider the fact that for longitudinal phonons the product $(\mathbf{P}\cdot\mathbf{e}_q)$ has the constant value, we can obtain after performing these calculations

$$\langle T_{\rm sol}\rangle_{\rm ph} = \exp\left\{-\frac{3{\rm j}MV_0^2\nu(\mathbf{P}\cdot\mathbf{e}_q)}{2\lambda_1 r_0\pi\hbar}{\rm th}(\nu(ir_0+Vt))\right\}. \quad (60)$$

Substituting equation (60) into equations (57, 58), we get

$$\langle T_{n\to m}\rangle = \overline{A}\sum_i(1+|\varphi_i|^2) \times\exp\left\{-\frac{3{\rm j}MV_0^2\nu(\mathbf{P}\cdot\mathbf{e}_q)}{2\lambda_1 r_0\pi h}{\rm th}(\nu(ir_0+Vt))\right\} \times{\rm e}^{{\rm j}i_0 r_0 p/\hbar}\exp\left\{-\sum_q\frac{(\mathbf{P}\cdot\mathbf{e}_q)}{2MN\hbar\omega_q}\left(\nu_q+\frac{1}{2}\right)\right\}. \quad (61)$$

In order to find out the explicit expression, we should calculate the sum in equation (61) over i. Thus we introduce the notation: $G = \frac{3MV_0^2\nu(\mathbf{P}\cdot\mathbf{e}_q)}{2\lambda_1 r_0\pi\hbar}$, and we make the continuum approximation: $\sum_i \to \frac{1}{r_0}\int_{-\infty}^{\infty}{\rm d}x$, so that the second term in the above expression becomes

$$\begin{aligned} F_1 &= \sum_i |\varphi_i|^2\exp\{-{\rm j}G{\rm th}(\nu(ir_0+Vt))\} \\ &= \frac{1}{r_0}\int_{-\infty}^{\infty}\frac{A^2(T){\rm d}x}{{\rm ch}^2(\nu(x-x_0-Vt))} \\ &\quad\times\exp\{-{\rm j}G{\rm th}(\nu(x+Vt))\}. \end{aligned}$$

Performing the corresponding transformation, we get approximately

$$\begin{aligned} F_1 &= \frac{A^2(T)}{T_0\nu}\exp\{-{\rm j}G{\rm th}(\nu(x_0+2Vt))\} \\ &\quad\times 2\int_0^1{\rm d}Y(1-Y^2)^0\cos(GY) \\ &= \frac{A^2(T)}{G}\exp\{-{\rm j}G{\rm th}(\nu(x_0+2Vt))\}\mathcal{J}_0(G) \end{aligned} \quad (62)$$

where $\mathcal{J}_0(G)$ is the zero order spherical Bessel function. Also, for the first term in equation (61) we obtain

$$F_2 = \sum_i\exp\{-{\rm j}G{\rm th}(\nu(ir_0+Vt))\}.$$

Using the continuum approximation $\sum_i \to \frac{1}{r_0}\int_{-\infty}^{\infty}{\rm d}x$, the above expression becomes approximately

$$\begin{aligned} F_2 &= \frac{1}{r_0}\int_{-\infty}^{\infty}\exp(-{\rm j}G{\rm th}(\nu x))\exp(-{\rm j}G{\rm th}(\nu Vt)){\rm d}x \\ &= \exp(-{\rm j}G{\rm th}(\nu Vt))\left(\frac{1}{r_0\nu}\right)\int_{-1}^{1}({\rm e}^{-{\rm j}GY}/{\rm sech}^2Y){\rm d}Y \\ &= \frac{2(G^2{\rm ch}^2 1+1)}{\nu G(G^2+1)}\exp\{-{\rm j}G{\rm th}(\nu Vt)\}. \end{aligned} \quad (63)$$

Thus equation (61) can be written as

$$\begin{aligned} \overline{\langle T_{n\to m}\rangle} &= (4\overline{A}/G)\left\{A^2(T)\mathcal{J}_0(G) \right. \\ &\quad\left.\times\exp(-{\rm j}G{\rm th}(\nu(Vt+x_0))) + \frac{(2(G^2{\rm ch}^2 1+1)}{\nu G(G^2+1)}\right\} \\ &\quad\times\exp(-{\rm j}G{\rm th}(\nu Vt)+{\rm j}r_0 i_0 P/\hbar) \\ &\quad\times\exp\left(-\sum_q\frac{(\mathbf{P}\cdot\mathbf{e}_q)}{2MN\omega_q\hbar}(\nu_q+1/2)\right). \end{aligned} \quad (64)$$

Finally, the Mössbauer transition probability is given as

$$\begin{aligned} &\tau = W_{n\to m} = |\langle T_{n\to m}\rangle|^2 \\ &= \frac{\lambda_1^2 r_0^2\pi^2\hbar^2\overline{A}^2}{9M^2V_0^4\nu^2(\mathbf{p}\cdot\mathbf{e}_q)^2}\{[A^2(T)\mathcal{J}_0(G)\cos(G{\rm th}(\nu(Vt+x_0))) \\ &+ \frac{2(G^2{\rm ch}^2 1+1)}{\nu G(G^2+1)}]^2 + \mathcal{J}_0(G)A^4(T)\sin^2(G{\rm th}(\nu(Vt+x_0))) \\ &\times\exp\left\{-\sum_q\frac{(\mathbf{p}\cdot\mathbf{e}_q)^2}{MN\hbar\omega_q}(\nu_q+\frac{1}{2})\right\}. \end{aligned} \quad (65)$$

We here are interested in the long-time behaviour of the transition probability in the steady situation, which is

$$\tau = \frac{\lambda_1^2 r_0^2 \pi^2 \hbar^2 \overline{A}^2}{9M^2 V_0^4 \nu^2 (\mathbf{p}\cdot\mathbf{e}_q)^2} \Big\{ \Big(A^2(T)\mathcal{J}_0(G)\cos G + \frac{2(G^2\mathrm{ch}^2 1+1)}{\nu(G^2+1)}\Big)^2 + A^4(T)\mathcal{J}_0^2(G)\sin^2 G\Big\} \times \exp\left\{-\sum_q \frac{(\mathbf{p}\cdot\mathbf{e}_q)^2}{MN\hbar\omega_q}\left(\nu_q+\frac{1}{2}\right)\right\}. \tag{66}$$

From equation (66) we can obtain the following properties of the Mössbauer effect arising from this mechanism we study here:

(1) the transition probability is a product of the supersonic soliton part resulting from the anharmonic effect at a finite temperature $T \neq 0$ K and the phononic part. If we admit that the molecular chain is populated exclusively by the solitons, the Mössbauer effect, generally speaking, is less probable than in the case where pure phonon modes are spread over the whole chain. The latter leads to the probability in equation (66) having the factor $\exp\left\{-\sum_q \frac{(\mathbf{P}\cdot\mathbf{e}_q)^2}{2MN\hbar\omega_q}\mathrm{cth}\left(\frac{\hbar\omega_q}{2K_\mathrm{B}T}\right)\right\}$.

(2) The above transition probability depends on the temperature T. As to the influence of the temperature on the probability, it is manifested through the factors $A(T)$, the amplitude of the soliton, and $\nu_q = [\exp(\hbar\omega_q/K_\mathrm{B}T) - 1]^{-1}$ in the case of pure phonon states. Knowledge of these parameters allows one to estimate the value of transition probability. In the general case, an accurate calculation for the probability is very difficult, but an approximate numerical estimation of equation (66) is possible by using generally accepted values for the parameters. In the literature [1–14,21] there is agreement on the following values: J = 3–4 cm^{-1}, $\chi = \frac{\hbar\chi_1}{2\omega_0}$ = (56–62) PN, $\chi' = \hbar\chi_2/2\omega_0$ = (6–8) PN, $M = 2.25 \times 10^{-25}$ kg, λ = (4.8–13) N/m, $\lambda_1 = (4 \times 10^{-5})$ N/m^2, r_0 = (2–5) Å, ε_0 = (0.2–0.205) eV, for acetanilide. Using the above values, in Figure 1 we plot the relative transition probability $\tau/\overline{A}^2$ *versus* the temperature T relation at $V = 1.2V_0$ and the wavelength of gamma-photon $\overline{\lambda} = 2.25 \times 10^{-11}$ m. From this figure we see that the transition probability increases slowly as the temperature increases in a linear-like fashion. However, the main effect of the temperature on the probability comes basically from the phononic part. Meanwhile, we can get that the $\tau/\overline{A}^2$ is 11–11.5% at T = 300 K and V/V_0 = 1.2. Therefore, the transition probability is very small at 300 K.

(3) The transition probability depends on the strength of the anharmonical vibration, λ_1. From equations (65, 66) we can see that the probability is zero, when the anharmonicity is not present. This shows clearly that the Mössbauer effect and corresponding transition probability comes from the anharmonical vibrations of molecular chains and the temperature in the systems. In Figure 2 we plot the τ^2/A^2 as a function of λ_1/λ at T = 300 K.

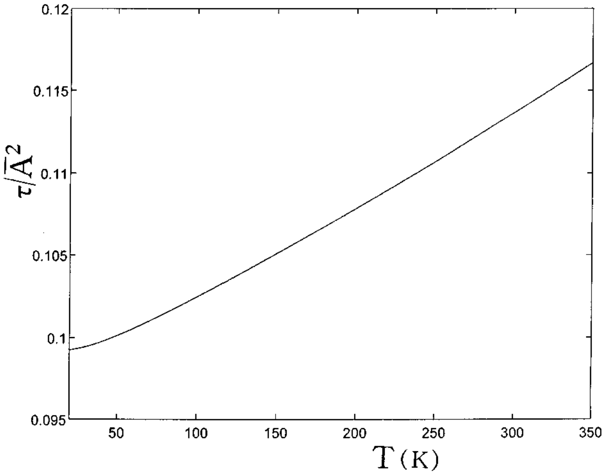

Fig. 1. The relative transition probability $\tau/\overline{A}^2$ *versus* the temperature T relation at $V = 1.2V_0$ and $\overline{\lambda} = 2.25 \times 10^{-11}$ m in equation (66).

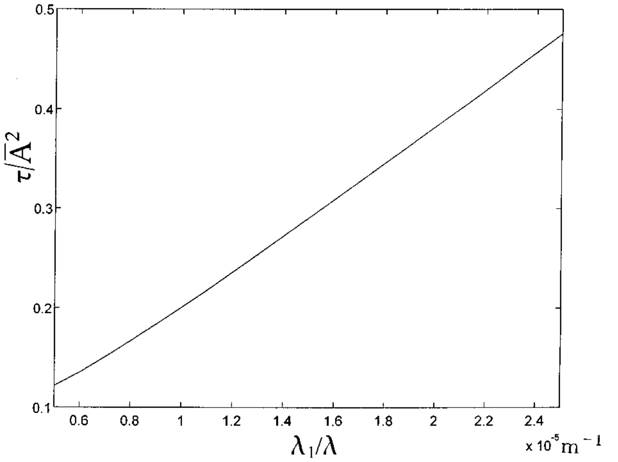

Fig. 2. The relative transition probability $\tau/\overline{A}^2$ *versus* the λ_1/λ relation at T = 300 K and $\overline{\lambda} = 2.25 \times 10^{-11}$ m in equation (66).

From this figure we see that $\tau/\overline{A}^2$ increases with increasing λ_1/λ.

(4) The transition probability depends on the strength of the coupling, $(\chi_1+\chi_2)$ and μ ($\mu = \nu r_0$). In Figures 3 and 4 we plot the $\tau/\overline{A}^2$ *versus* the $(\chi_1+\chi_2)$ and μ relations, respectively. From the two figures we see that the $\tau/\overline{A}^2$ increases with increasing the $(\chi_1+\chi_2)$ and decreasing the μ at V/V_0 =1.2, T =300 K and $\overline{\lambda} = 2.25\times10^{-11}$ m, respectively. From equation (66) we can also see that the transition probability is pure phononic, when the $(\chi_1+\chi_2) = 0$. This shows clearly that this Mössbauer effect and corresponding transition probability result from the motions of the solitons and the thermal phonons, and their interaction in the organic molecular crystals. Also, the transition probability decreases with growth of the soliton velocity, V, since the parameter $\mu = \left[\frac{1}{2}(V^2/V_0^2-1)\right]^{1/2}$ is related to V.

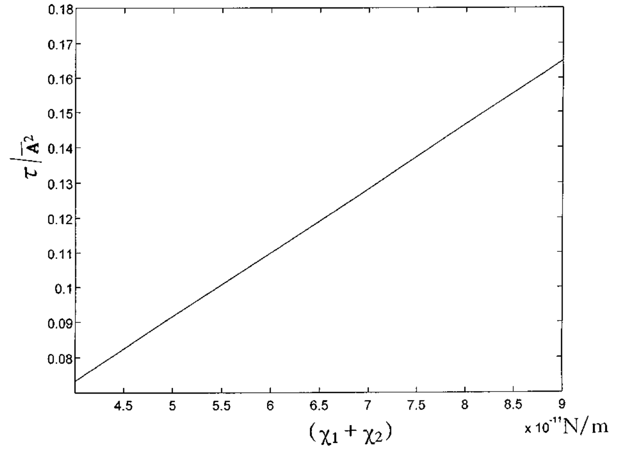

Fig. 3. The relative transition probability $\tau/\overline{A}^2$ *versus* the $(\chi_1 + \chi_2)$ relation at $V = 1.2V_0$, $T = 300$ K and $\overline{\lambda} = 2.25 \times 10^{-11}$ m in equation (66).

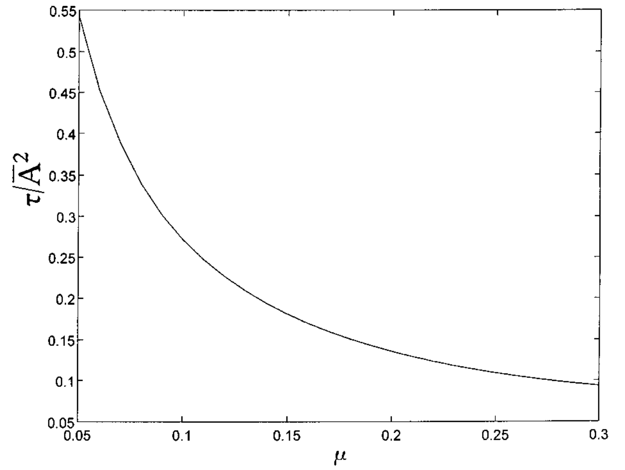

Fig. 4. The relative transition probability $\tau/\overline{A}^2$ *versus* the μ at $V = 1.2V_0$ and $\overline{\lambda} = 2.25 \times 10^{-11}$ m and $T = 300$ K in equation (66).

(5) The transition probability is related to the wavelength of the γ-photon emitted. In Figure 5 we plot $\tau/\overline{A}^2$ *versus* $\overline{\lambda}$ relation. From this figure we see the $\tau/\overline{A}^2$ is smaller when the wavelength $\overline{\lambda}$ is shorter.

(6) The size of the transition probabilities depends on the inherent characteristics of organic molecular crystals $(M, r_0, V_0, N, \omega_0$ and $\omega_1)$, and the features of these quasi-particles generated in collective excitation $(\omega_q, q, V$ and $m)$, and the environment condition of molecular crystals, *i.e.*, the temperature. Therefore, the transition probability for different molecular crystals is different.

If we suppose that the scalar product $(\mathbf{P} \cdot \mathbf{e})$ is small enough so that the estimation $G \ll 1$ is valid, then in such a case the transition probability becomes

$$W_{n \to m}(G \to 0) \propto (1 + 3/8(s^2 - 1) \times \exp\left(-\sum_q \left(\frac{(\mathbf{p} \cdot \mathbf{e}_q)^2}{2MN\hbar\omega_q}\right) \operatorname{cth}\left(\frac{\hbar V_0 q}{2K_\mathrm{B}T}\right)\right).$$

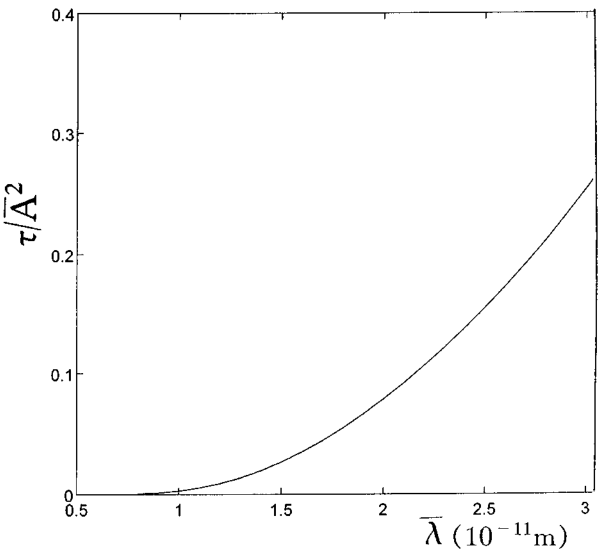

Fig. 5. The relative transition probability $\tau/\overline{A}^2$ *versus* the wavelength of ν-photon, $\overline{\lambda}$ relation at $V = 1.2V_0$ and $T = 300$ K in equation (66).

On the other hand, for large values of G we obtain approximately

$$W_{n \to m}(G \to \infty) \approx \overline{A}^2 \left(\frac{\lambda_1 r_0^2 \hbar}{MV_0^2(\mathbf{p} \cdot \mathbf{e}_q)}\right)^4 A^2(T)(\pi/\nu)(A^2(T)\cos^2 G + \sin^2 G) \times \exp\left(-\sum_q \left(\frac{(\mathbf{p} \cdot \mathbf{e}_q)^2}{2MN\hbar\omega_q}\right) \operatorname{cth}\left(\frac{\hbar V_0 q}{2K_\mathrm{B}T}\right)\right)$$

where the probability rapidly decreases with the increase of parameter G so that the solitonic Mössbauer effect is basically negligible in such a case.

The results obtained above develop the subject of the soliton model of the intramolecular energy transport in molecular crystals, and facilitate further the experimental confirmation of the soliton existence in the molecular crystals by using the Mössbauer effect method. This study could also advance development of soliton physics and Mössbauer technology.

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