# Solving the initial condition of the string relaxation equation of the string model for glass transition: part-I*

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The string model for the glass transition can quantitatively describe the universal $\alpha$-relaxation in glassformers, including the average relaxation time, the distribution function of the relaxation time, and the relaxation strength as functions of temperature. The string relaxation equation (SRE) of the model, at high enough temperatures, simplifies to the well-known single particle mean-field Debye relaxation equation, and at low enough temperatures to the well-known Rouse–Zimm relaxation equation that describes the relaxation dynamics of linear macromolecules. However, its initial condition, necessary to the further model predictions of glassy dynamics, has not been solved. In this paper, the special initial condition (SIC) of the SRE, i.e. for straight strings and the dielectric spectrum technique that is one of the most common methods to measure the glassy dynamics, was solved exactly. It should be expected that the obtained SIC would benefit the solution of the general initial condition of the SRE of the string model, i.e. for stochastically spatially configurating strings, as will be described in separate publications.

**Keywords:** glass transition, relaxation phenomenon, dielectric relaxation

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## 1. Introduction

Though the glass transition is a fundamental property of the condensed matter, none of the mechanisms and models proposed to explain this property have widely received acceptance, remaining a central unresolved issue in condensed matter physics.[1−60] Recently, a molecular level mechanism of coupling dynamical molecule strings was proposed by Huang *et al.* called the string model for the sake of convenience.[19] Besides describing the single molecular motions of Debye's mean-field theory approximation,[20] the string model represents the collective units of the structure and dynamics in glassformers by coupling molecular strings randomly distributed in space, characteristics which are widely observed in well-designed experiments, analog simulations and molecular dynamics simulations.[21−23] The reasons we think that the basic structural and dynamical units of liquid state being the dynamical molecular string rather than the dynamical branched clusters are: 1) formally, supposing to first 'walk' along a branch of a cluster, and if we meet the cross point of two or more branches, then we will randomly choose a branch to 'walk', meanwhile 'cut' the other branches. By doing such process repeatedly, the cluster will formally be divided into 'individual' strings; 2) physically, the above processes are to re-express the cluster by some interacting strings, and the inter-string interacting points are the cross points of the branches; 3) mathematically, the above processes bring the corresponding mathematical problems become more easy to solve. Furthermore, the model assumes relative weak interactions between the strings, compared with the intra-string ones.

In liquids, Debye's theory[20] describes the mean-field of the orientational and translational motions of the $i$th molecule by the well-known sin-Gordon potential,

$$\phi_i^{\mathrm{sG}} = \frac{V_0}{2}\left[1-\cos(2(\theta_i-\theta_i^0))\right]\delta_{\varphi_i,\varphi_i^0},$$

and the $\phi^{(4)}$ potential,

$$\phi_i^{(4)} = 4V_0\Big[-\frac{1}{2}\left(\frac{r_i-r_i^0}{d}\right)^2$$

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$$+\frac{1}{4}\left(\frac{r_i-r_i^0}{d}\right)^4\Bigg]\delta_{\theta_i,\theta_i^0}\delta_{\varphi_i,\varphi_i^0},$$

respectively. Here $V_0$ and $d$ are the barrier height and width in the double-wells; $\theta_i^0$ and $\varphi_i^0$ are the directional and rotational angles of the double-wells in space, respectively, and they symmetrically distribute in the range $[0,\pi]$ and $[0,2\pi]$. And $r_i(0\leq r_i<\infty)$, $\theta_i$ $(0\leq\theta_i<\pi)$, and $\varphi_i$ $(0\leq\varphi_i<2\pi)$ are, respectively, the radius vector, directional and rotational angles of the $i$th molecule; and $\delta_{\theta_i,\theta_i^0}$ and $\delta_{\varphi_i,\varphi_i^0}$ are the Kronig-$\delta$ function.

Furthermore, due to the fact that $\mathrm{e}^{-V_0/k_\mathrm{B}T}\ll 1$ in liquids ($k_\mathrm{B}$ is the Boltzmann's constant, and $T$ the absolute temperature), molecular jumping processes between the double wells are equivalent to those between two states, $\sigma_i=\pm1$, approximately, at a jumping rate $v_0\mathrm{e}^{-V_0/k_\mathrm{B}T}$ ($\nu_0$ is the vibration frequency of molecules in the wells).

Taking into account above principles, the string model represents intra-string interactions by using a finite one-dimensional Ising model $H_1=-V\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{n-1}\sigma_i^{mn}\sigma_{i+1}^{mn}$,[20] whereas the inter-string interactions are represented by a random Ising interacting model

$$H_2=\frac{V}{2}\sum_{m=1}^{M}\sum_{i=1}^{n}\sum_{m'\neq m,i'=1}^{nn(i)}\sigma_i^{mn}\sigma_{i'}^{m'n'}\Theta_{ii'}^{mm'},$$

where $V$ is the corresponding interaction constant, independent of temperature. The symbol $\sigma_i^{mn}=\pm1$ denotes the two states of the $i$th molecule of a string (numbered $m$ in the system) containing $n$ molecules (called n-string hereafter), $nn(k)$ represents the nearest number of molecules surrounding the $i$th molecule, $\Theta_{ii'}^{mm'}$ is a random distribution function in the range $[-1,1]$, and $M$ is the total number of molecule strings in a liquid.

So, the string model describes the coupled orientational or translational jumping motions of molecules in liquids, supercooled liquids and the glass by

$$H_\mathrm{e}=-V\sum_{m=1}^{M}\left(\sum_{i=1}^{n-1}\sigma_i^{mn}\sigma_{i+1}^{mn}-\frac{1}{2}\sum_{i=1}^{n}\sum_{m'\neq m,i'=1}^{nn(i)}\sigma_i^{mn}\sigma_{i'}^{m'n'}\Theta_{ii'}^{mm'}\right),$$
$$\tau_0=\nu_0^{-1}\mathrm{e}^{V_0/k_\mathrm{B}T}. \qquad (1)$$

This equation is the Hamiltonian of a partly random Ising model[20] in a definite jumping time.

The main conclusions of the model for a glassy system under a first-order approximation (the calculation error is estimated to be about 1% near the glass transition temperature $T_\mathrm{g}$) are:[19] 1) the relaxation behaviour of a molecular string is equivalent to that of an effective molecule (EM), characterized by a definite relaxation time and dipole moment; 2) an ordinary liquid at high temperature is renormalised to an EM gas, this approach presenting a possible brand-new kind of picture about the ordinary liquid; 3) the strong correlated supercooled liquid at low temperatures is renormalised to a weak correlated EM liquid; and 4) there exists a crossover transition between the EM gas and the EM liquid.

The polarisability of the system predicted by the model gives a unified and quantitative description of the $\alpha$-relaxation dynamics in glass formers including: i) the relaxation time crossover from high temperature Arrhenius behaviour to low temperature Vogel–Fulcher–Tammann law;[61]; ii) the crossover of the relaxation function from a high temperature Debye exponential to a low temperature Kohlrausch–Williams–Watts[62] or Cole–Davidson[63] functions; and iii) the departure of the relaxation intensity from the Curie law.[16−18]

In addition, compared with other models, least parameters of clear physical origin are involved in the string model.[19]

The physical core of the string model arises from the intra-string interaction $H_1$ described by Eq. (1), which leads to the following relaxation equations for an individual $n$-string,[19]

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix}\delta_1\\ \delta_2\\ \vdots\\ \vdots\\ \vdots\\ \delta_{n-1}\\ \delta_n\end{bmatrix} = -\nu_0 \mathrm{e}^{-V_0/kT}\begin{bmatrix} 1 & 2u-1 & 0 & \cdots & 0 & 0 & 0\\ w-1/2 & 1 & w-1/2 & \cdots & \vdots & 0 & 0\\ \vdots & w-1/2 & 1 & \cdots & \vdots & \vdots & 0\\ \vdots & \vdots & w-1/2 & \cdots & w-1/2 & \vdots & \vdots\\ 0 & \vdots & \vdots & \cdots & 1 & w-1/2 & \vdots\\ 0 & 0 & \vdots & \cdots & w-1/2 & 1 & w-1/2\\ 0 & 0 & 0 & \cdots & 0 & 2u-1 & 1 \end{bmatrix}\begin{bmatrix}\delta_1\\ \delta_2\\ \vdots\\ \vdots\\ \vdots\\ \delta_{n-1}\\ \delta_n\end{bmatrix}, \quad (2)$$

where $\delta_i^{(n)}(t) \equiv p_i^{(n)}(t) - p_i^{(n)}(\infty)$, $i = 1, \ldots, n$. The $p_i^{(n)}(t)$ is the probability that the $i$th molecule of an n-string is at the orientational state $\sigma_i^{mn} = 1$, and $p_i^{(n)}(\infty)$ is the relevant probability at equilibrium. Therefore, $\delta_i^{(n)}(t)$ is the departure of the property from equilibrium; $u \equiv \left(1 + \mathrm{e}^{2v}\right)^{-1}$ and $w \equiv \left(1 + \mathrm{e}^{4v}\right)^{-1}$.

For the sake of convenience, Eq. (2) will be called the string relaxation equation (SRE), whereas the square matrix on the right side of Eq. (2) will be named $M_{\mathrm{SR}}$. At high enough temperatures, i.e. $v \to 0$, $M_{\mathrm{SR}}$ becomes a unit matrix and the SRE simplifies to the well-known Debye relaxation equation of single particle mean-field.[20] On the other hand, at low enough temperature, i.e. $v \to \infty$, and $M_{\mathrm{SR}}$ is the Rouse–Zimm matrix, the SRE becoming the well-known Rouse–Zimm relaxation equation that describes the relaxation dynamics of linear macromolecules.[64] Since the Debye theory and the Rouse model are the two most successful relaxation theories for monomers and macromolecules, respectively, the string model could be considered, at least mathematically a universal model that describes the relaxation dynamics of the amorphous condensed matter.

Physically, the relaxation of the $n$ coupled molecules in an $n$-string is described by $n$ individual relaxation modes, according to the SRE. The first-order approximation in Ref. [19] only gives the main relaxation mode corresponding to the largest relaxation intensity and the longest relaxation time, the other modes being omitted. In addition to the $\alpha$-relaxation, glass formers exhibit a universal well-known Johari–Goldstein $\beta$-relaxation at high frequencies or low temperatures.[16−18] The microscopic mechanism at molecular level associated with the Johari–Goldstein $\beta$-relaxation is not clear till now, though there are some phenomenological theories, such as the coupling model[10] and the mode-coupling theory,[11] which give a glimpse of the characteristics of this process. The relaxation mode scenario of the string model possibly provides a unified mechanism of the $\alpha$- and $\beta$-relaxations. It can be assumed that with the exception of the main relaxation mode associated to the $\alpha$-relaxation, all the other modes correspond to the Johari–Goldstein $\beta$-process. To carry out the exact analysis and quantitative comparison of these assumptions with experiments, it is necessary to calculate the strengths of all the modes. To accomplish this goal, it is necessary to solve the initial conditions of the SRE, that is,

$$\delta_i^{(n)}(0) \equiv p_i^{(n)}(0) - p_i^{(n)}(\infty), \quad i = 1, \ldots, n. \quad (3)$$

Owing to the stochastic spatial configurations of the molecular strings, the general solution of the initial condition involves serious difficulties. Because of this, in what follows, we only study the case of straight molecular strings, a special case, called the special initial condition (SIC) of the SRE.

## 2. General calculation method

Similar to the method described in Ref. [19], the relevant question of the motion of n molecules coupled in an $n$-string is first transformed to that of the individual $2^n$ orientational states. The motion of each one of these $2^n$ states can be exactly calculated using the Boltzmann principle, and the results thus obtained are transformed back to those of the coupled molecules.

Although the SRE is independent of experimental methods in the linear response regime, its initial condition shown as Eq. (3) depends. In Ref. [19], Huang *et al.* have calculated the permittivity of glassformers because this property provides the glassy dynamics in a wide frequency range.[16−18] In this technique, the relevant SIC of an $n$-string corresponding to $\delta_i^{(n)}(0)$,

$i = 1, \ldots, n$ is induced by an external electric field. Without losing generality, and in the linear response regime,[20] let us consider an $n$-string perturbed by a small enough step electric field along the string,

$$F(t) = \begin{cases} F_0, & t < 0; \\ 0, & t \geq 0. \end{cases} \quad (4)$$

Mathematically, the linear response condition is that the reduced energy of the molecule in the electric field $f \equiv \mu_0 F_0 / k_{\rm B} T \ll 1$. In this expression, $\mu_0$ is the permanent electric dipole moment of the molecule along the string direction.

## 3. Direct calculation method of SIC and questions

Since a molecule in the string model has two possible orientations, an $n$-string has $2^n$ orientational states. Let $E_j^{(n)}(F)$ and $q_j^{(n)}(t)$ be, respectively, the energy and the probability of the $j$th orientational state of the $n$-string in the electric field. According to the Boltzmann principle,

$$q_j^{(n)}(0) = {\rm e}^{-E_j^{(n)}(F_0)/kT} \Big/ \sum_{k=1}^{2^n} {\rm e}^{-E_k^{(n)}(F_0)/kT},$$
$$q_j^{(n)}(\infty) = {\rm e}^{-E_j^{(n)}(0)/kT} \Big/ \sum_{k=1}^{2^n} {\rm e}^{-E_k^{(n)}(0)/kT}. \quad (5)$$

As an example, the $2^4$ orientational states of a 4-string are shown in Table 1, where $Q_n = 2\,(2{\rm ch}v)^{n-1}$ is the orientational partition function of the $n$-string,[19] and ${\rm ch}v \equiv ({\rm e}^v + {\rm e}^{-v})/2$.

Since the two orientations $\sigma_i^{mn} = 1$ and $\sigma_i^{mn} = -1$ of a molecule can be formally expressed by '0' and '1', respectively, the total orientational states of an n-string can be denoted by n binary digits, as shown in the 2nd columns of Table 1. For example, '0000' indicates that $\sigma_i^{m4} = 1, i = 1, 2, 3, 4$. The 3rd and 4th columns denote $E_j^{(4)}(0)$ and $E_j^{(4)}(F_0)$ for the $j$th orientational state when the external field is 0 and $F_0$, respectively, whereas the relevant quantities $q_j^{(4)}(\infty)$ and $q_j^{(4)}(0)$ for $t \to \infty$ and $t = 0$, respectively, are shown in the 5th, 6th columns.

$E_j^{(n)}(F)$ has two parts, one originating from the intra-string interaction $H_1$ of Eq. (1) and the other from the permanent electric dipole moment $\mu_0$ of the molecule in the electric field, e.g. for the '0000' orientational state, the intra-string energy and the energy of string molecules in the electric field are $-3V$ and $-4\mu_0 F$, respectively. For the linear response case $f \ll 1$ and ${\rm e}^f \approx 1 + f$, as shown in the 6th column of the Table 1.

**Table 1.** Expressions for the total orientational states of a straight 4-string and the relevant quantities in the external electric field of Eq. (4).

| serial number | orientational states | $E_j^{(4)}(0)$ | $E_j^{(4)}(F_0)$ | $q_j^{(4)}(\infty)Q_4$ | $q_j^{(4)}(0)Q_4$ |
|---|---|---|---|---|---|
| $j$=1 | 0000 | $-3V$ | $-3V - 4\mu_0 F_0$ | ${\rm e}^{3v}$ | $(1+4f)\,{\rm e}^{3v}$ |
| $j$=2 | 0001 | $-V$ | $-V - 2\mu_0 F_0$ | ${\rm e}^{v}$ | $(1+2f)\,{\rm e}^{v}$ |
| $j$=3 | 0010 | $V$ | $V - 2\mu_0 F_0$ | ${\rm e}^{-v}$ | $(1+2f)\,{\rm e}^{-v}$ |
| $j$=4 | 0011 | $-V$ | $-V$ | ${\rm e}^{v}$ | ${\rm e}^{v}$ |
| $j$=5 | 0100 | $V$ | $V - 2\mu_0 F_0$ | ${\rm e}^{-v}$ | $(1+2f)\,{\rm e}^{-v}$ |
| $j$=6 | 0101 | $3V$ | $3V$ | ${\rm e}^{-3v}$ | ${\rm e}^{-3v}$ |
| $j$=7 | 0110 | $V$ | $V$ | ${\rm e}^{-v}$ | ${\rm e}^{-v}$ |
| $j$=8 | 0111 | $-V$ | $-V + 2\mu_0 F_0$ | ${\rm e}^{v}$ | $(1-2f)\,{\rm e}^{v}$ |
| $j$=9 | 1000 | $-V$ | $-V - 2\mu_0 F_0$ | ${\rm e}^{v}$ | $(1+2f)\,{\rm e}^{v}$ |
| $j$=10 | 1001 | $V$ | $V$ | ${\rm e}^{-v}$ | ${\rm e}^{-v}$ |
| $j$=11 | 1010 | $3V$ | $3V$ | ${\rm e}^{-3v}$ | ${\rm e}^{-3v}$ |
| $j$=12 | 1011 | $V$ | $V + 2\mu_0 F_0$ | ${\rm e}^{-v}$ | $(1-2f)\,{\rm e}^{-v}$ |
| $j$=13 | 1100 | $-V$ | $-V$ | ${\rm e}^{v}$ | ${\rm e}^{v}$ |
| $j$=14 | 1101 | $V$ | $V + 2\mu_0 F_0$ | ${\rm e}^{-v}$ | $(1-2f)\,{\rm e}^{-v}$ |
| $j$=15 | 1110 | $-V$ | $-V + 2\mu_0 F_0$ | ${\rm e}^{v}$ | $(1-2f)\,{\rm e}^{v}$ |
| $j$=16 | 1111 | $-3V$ | $-3V + 4\mu_0 F_0$ | ${\rm e}^{3v}$ | $(1-4f)\,{\rm e}^{3v}$ |

From the definition given for $p_i^{(n)}(t)$ above and taking into account Table 1, the following relationships between $p_i^{(4)}(t)$ and $q_j^{(4)}(t)$ are obtained,

$$
\begin{aligned}
p_1^{(4)}(t) &= q_1^{(4)}(t) + q_2^{(4)}(t) + q_3^{(4)}(t) + q_4^{(4)}(t) + q_5^{(4)}(t) + q_6^{(4)}(t) + q_7^{(4)}(t) + q_8^{(4)}(t),\\
p_2^{(4)}(t) &= q_1^{(4)}(t) + q_2^{(4)}(t) + q_3^{(4)}(t) + q_4^{(4)}(t) + q_9^{(4)}(t) + q_{10}^{(4)}(t) + q_{11}^{(4)}(t) + q_{12}^{(4)}(t),\\
p_3^{(4)}(t) &= q_1^{(4)}(t) + q_2^{(4)}(t) + q_5^{(4)}(t) + q_5^{(4)}(t) + q_9^{(4)}(t) + q_{10}^{(4)}(t) + q_{13}^{(4)}(t) + q_{14}^{(4)}(t),\\
p_4^{(4)}(t) &= q_1^{(4)}(t) + q_3^{(4)}(t) + q_5^{(4)}(t) + q_7^{(4)}(t) + q_9^{(4)}(t) + q_{11}^{(4)}(t) + q_{13}^{(4)}(t) + q_{15}^{(4)}(t). \qquad (6)
\end{aligned}
$$

The specific method for the calculation is first to find all the orientational states with the first (from left to right) binary digit being '0', as shown in Table 1, and then to add them. Thus $\delta_i^{(4)}(0), i = 1, \ldots, 4$ can exactly be calculated by means of Eqs. (3) and (6) as well as the results of $q_j^{(4)}(0)$ and $q_j^{(4)}(\infty)$ presented in Table 1. This procedure is called the direct calculation method.

For an arbitrary string length, the SIC can be calculated by the same method as that used for the 4-string, and the pertinent results for $n = 1$ to 7 are shown in Table 2. The results in the table indicate that $\delta_i^{(n)}(0) = \delta_{n-i+1}^{(n)}(0)$, $i = 1, \ldots, n$, an equality which could be forecasted from the symmetry of the straight string.

**Table 2.** Strict solution of the SIC for $n = 1$ to 7 in the external field of Eq. (4) using the direct calculation method.

| $n$ | $\delta_1^{(n)}(0)Q_n/f$ | $\delta_2^{(n)}(0)Q_n/f$ | $\delta_3^{(n)}(0)Q_n/f$ | $\delta_4^{(n)}(0)Q_n/f$ |
|---|---|---|---|---|
| 1 | 1 | | | |
| 2 | $2e^{v}$ | | | |
| 3 | $3e^{2v} + e^{-2v}$ | $3e^{2v} + 2 - e^{-2v}$ | | |
| 4 | $4e^{3v} + 4e^{-v}$ | $4e^{3v} + 4e^{v}$ | | |
| 5 | $5e^{4v} + 10 + e^{-4v}$ | $5e^{4v} + 6e^{2v} + 4 + 2e^{-2v} - e^{-4v}$ | $5e^{4v} + 8e^{2v} + 2 + e^{-4v}$ | |
| 6 | $6e^{5v} + 20e^{v} + 6e^{-3v}$ | $6e^{5v} + 8e^{3v} + 12e^{v} + 8e^{-v} - 2e^{-3v}$ | $6e^{5v} + 12e^{3v} + 8e^{v} + 4e^{-v} + 2e^{-3v}$ | |
| 7 | $7e^{6v} + 36e^{2v} + 20e^{-4v}$ $+e^{-6v}$ | $7e^{6v} + 10e^{4v} + 25e^{2v} + 20$ $+2e^{-2v} + 2e^{-4v} - e^{-6v}$ | $7e^{6v} + 10e^{4v} + 25e^{2v} + 20$ $+2e^{-2v} + 2e^{-4v} - e^{-6v}$ | $7e^{6v} + 18e^{4v} + 18e^{2v} + 18$ $+8e^{-2v} + 2e^{-4v} - e^{-6v}$ |

An inspection of the results in Table 2 shows that with increasing n, the expressions for $\delta_i^{(n)}(0)$ become more and more complex. Specifically, the number of terms for $\delta_i^{(n)}(0)$ increases rapidly and the exponential factors of each term become higher and higher. Unfortunately, except for the first term, the regularity of $\delta_i^{(n)}(0)$ for arbitrary values of n is not found. We would like to point out that for n $\sim$ 50, the number of total orientational states of an $n$-string is roughly $2^{50}$ around the glass transition temperature.[19] Then the use of the direct calculation method looks not practically feasible.

## 4. Recursive calculation method for SIC and results

This section describes a recursive method to calculate $\delta_i^{(n)}(0)$. The key point of the method is to relate the energy of $j^{\text{th}}$ orientational state of the $n$-string $E_j^{(n)}(F)$, to the energies of shorter strings $E_k^{(n-1)}(F)$, $E_l^{(n-2)}(F), \ldots, E_1^{(1)}(F)$. Then the recursive relations between $q_j^{(n)}(t)$ and $q_k^{(n-1)}(t)$, $q_l^{(n-2)}(t), \ldots q_1^{(1)}(t)$, and consequently the recursive relations between $p_i^{(n)}(0)$ of the $n$-string and $p_k^{(n-1)}(t)$, $p_l^{(n-2)}(t), \ldots p_1^{(1)}(t)$ of shorter strings are found. Finally, the values of $\delta_i^{(n)}(0), i = 1, \ldots, n$ are calculated using Eq. (3). In what follows, the 4-string is utilized as an example to calculate $p_i^{(n)}(\infty)$. The specific calculation processes are

$$
\begin{aligned}
p_1^{(4)}(\infty) &= q_1^{(4)}(\infty) + q_2^{(4)}(\infty) + q_3^{(4)}(\infty) + q_4^{(4)}(\infty) + q_5^{(4)}(\infty) + q_6^{(4)}(\infty) + q_7^{(4)}(\infty) + q_8^{(4)}(\infty)\\
&= \left[e^{-e_1^{(4)}(0)} + e^{-e_2^{(4)}(0)} + e^{-e_3^{(4)}(0)} + e^{-e_4^{(4)}(0)} + e^{-e_5^{(4)}(0)} + e^{-e_6^{(4)}(0)} + e^{-e_7^{(4)}(0)} + e^{-e_8^{(4)}(0)}\right]/Q_4\\
&= \left[e^{-e_1^{(3)}(0)+v} + e^{-e_2^{(3)}(0)+v} + e^{-e_3^{(3)}(0)+v} + e^{-e_4^{(3)}(0)+v} + e^{-e_5^{(3)}(0)-v} + e^{-e_6^{(3)}(0)-v} + e^{-e_7^{(3)}(0)-v} + e^{-e_8^{(3)}(0)-v}\right]/Q_4\\
&= \left[\left(q_1^{(3)}(\infty) + q_2^{(3)}(\infty) + q_3^{(3)}(\infty) + q_4^{(3)}(\infty)\right)e^{v} + \left(q_5^{(2)}(\infty) + q_6^{(2)}(\infty) + q_7^{(3)}(\infty) + q_8^{(3)}(\infty)\right)e^{-v}\right]Q_3/Q_4\\
&= \left[p_1^{(3)}(\infty)e^{v} + \left(1 - p_1^{(3)}(\infty)\right)e^{-v}\right]Q_3/Q_4,
\end{aligned}
$$

$$
\begin{aligned}
p_2^{(4)}(\infty) &= q_1^{(4)}(\infty)+q_2^{(4)}(\infty)+q_3^{(4)}(\infty)+q_4^{(4)}(\infty)+q_9^{(4)}(\infty)+q_{10}^{(4)}(\infty)+q_{11}^{(4)}(\infty)+q_{12}^{(4)}(\infty)\\
&= \left[\mathrm{e}^{-e_1^{(4)}(0)}+\mathrm{e}^{-e_2^{(4)}(0)}+\mathrm{e}^{-e_3^{(4)}(0)}+\mathrm{e}^{-e_4^{(4)}(0)}+\mathrm{e}^{-e_9^{(4)}(0)}+\mathrm{e}^{-e_{10}^{(4)}(0)}+\mathrm{e}^{-e_{11}^{(4)}(0)}+\mathrm{e}^{-e_{12}^{(4)}(0)}\right]\Big/Q_4\\
&= \left[\mathrm{e}^{-e_1^{(3)}(0)+v}+\mathrm{e}^{-e_2^{(3)}(0)+v}+\mathrm{e}^{-e_3^{(3)}(0)+v}+\mathrm{e}^{-e_4^{(3)}(0)+v}+\mathrm{e}^{-e_1^{(3)}(0)-v}+\mathrm{e}^{-e_2^{(3)}(0)-v}+\mathrm{e}^{-e_3^{(3)}(0)-v}+\mathrm{e}^{-e_4^{(3)}(0)-v}\right]\Big/Q_4,\\
&= \left(q_1^{(3)}(\infty)+q_2^{(3)}(\infty)+q_3^{(3)}(\infty)+q_4^{(3)}(\infty)\right)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_3/Q_4\\
&= p_1^{(3)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_3/Q_4,\\
p_3^{(4)}(\infty) &= q_1^{(4)}(\infty)+q_2^{(4)}(\infty)+q_5^{(4)}(\infty)+q_6^{(4)}(\infty)+q_9^{(4)}(\infty)+q_{10}^{(4)}(\infty)+q_{13}^{(4)}(\infty)+q_{14}^{(4)}(\infty)\\
&= \left[\mathrm{e}^{-e_1^{(4)}(0)}+\mathrm{e}^{-e_2^{(4)}(0)}+\mathrm{e}^{-e_5^{(4)}(0)}+\mathrm{e}^{-e_6^{(4)}(0)}+\mathrm{e}^{-e_9^{(4)}(0)}+\mathrm{e}^{-e_{10}^{(4)}(0)}+\mathrm{e}^{-e_{13}^{(4)}(0)}+\mathrm{e}^{-e_{14}^{(4)}(0)}\right]\Big/Q_4\\
&= \left[\mathrm{e}^{-e_1^{(3)}(0)+v}+\mathrm{e}^{-e_2^{(3)}(0)+v}+\mathrm{e}^{-e_5^{(3)}(0)-v}+\mathrm{e}^{-e_6^{(3)}(0)-v}+\mathrm{e}^{-e_1^{(3)}(0)-v}+\mathrm{e}^{-e_2^{(3)}(0)-v}+\mathrm{e}^{-e_5^{(3)}(0)+v}+\mathrm{e}^{-e_5^{(3)}(0)+v}\right]\Big/Q_4\\
&= \left(q_1^{(3)}(\infty)+q_2^{(3)}(\infty)+q_5^{(3)}(\infty)+q_6^{(3)}(\infty)\right)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_3/Q_4 = p_2^{(3)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_3/Q_4,\\
p_4^{(4)}(\infty) &= q_1^{(4)}(\infty)+q_3^{(4)}(\infty)+q_5^{(4)}(\infty)+q_7^{(4)}(\infty)+q_9^{(4)}(\infty)+q_{11}^{(4)}(\infty)+q_{13}^{(4)}(\infty)+q_{15}^{(4)}(\infty)\\
&= \left[\mathrm{e}^{-e_1^{(4)}(0)}+\mathrm{e}^{-e_3^{(4)}(0)}+\mathrm{e}^{-e_5^{(4)}(0)}+\mathrm{e}^{-e_7^{(4)}(0)}+\mathrm{e}^{-e_9^{(4)}(0)}+\mathrm{e}^{-e_{11}^{(4)}(0)}+\mathrm{e}^{-e_{13}^{(4)}(0)}+\mathrm{e}^{-e_{15}^{(4)}(0)}\right]\Big/Q_4\\
&= \left[\mathrm{e}^{-e_1^{(3)}(0)+v}+\mathrm{e}^{-e_3^{(3)}(0)+v}+\mathrm{e}^{-e_5^{(3)}(0)-v}+\mathrm{e}^{-e_7^{(3)}(0)-v}+\mathrm{e}^{-e_1^{(3)}(0)-v}+\mathrm{e}^{-e_3^{(3)}(0)-v}+\mathrm{e}^{-e_5^{(3)}(0)+v}+\mathrm{e}^{-e_7^{(3)}(0)+v}\right]\Big/Q_4\\
&= \left(q_1^{(3)}(\infty)+q_3^{(3)}(\infty)+q_5^{(3)}(\infty)+q_7^{(3)}(\infty)\right)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_3/Q_4\\
&= p_3^{(3)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_3/Q_4,
\end{aligned}
$$

where $e_j^{(n)}(F)\equiv E_j^{(n)}(F)/k_{\mathrm{B}}T$; whereas the expressions for $p_i^{(n)}(\infty)$ of the $n$-string ($n=1$ to 4) are shown in Table 3.

**Table 3.** The $p_i^{(n)}(\infty)$ of the $n$-string for $n=1$ to 4 in the external field of Eq. (4).

| $n$ | $p_1^{(n)}(\infty)Q_n/Q_{n-1}$ | $p_2^{(n)}(\infty)Q_n/Q_{n-1}$ | $p_3^{(n)}(\infty)Q_n/Q_{n-1}$ | $p_4^{(n)}(\infty)Q_n/Q_{n-1}$ |
|---|---|---|---|---|
| 1 | 1/2 | | | |
| 2 | $p_1^{(1)}(\infty)\mathrm{e}^{v}+\left(1-p_1^{(1)}(\infty)\right)\mathrm{e}^{-v}$ | $p_1^{(1)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)$ | | |
| 3 | $p_1^{(2)}(\infty)\mathrm{e}^{v}+\left(1-p_1^{(2)}(\infty)\right)\mathrm{e}^{-v}$ | $p_1^{(2)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)$ | $p_2^{(2)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)$ | |
| 4 | $p_1^{(3)}(\infty)\mathrm{e}^{v}+\left(1-p_1^{(3)}(\infty)\right)\mathrm{e}^{-v}$ | $p_1^{(3)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)$ | $p_2^{(3)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)$ | $p_3^{(3)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)$ |

From Table 3, the recursive relation of $p_i^{(n)}(\infty)$ for arbitrary $n$ is

$$
\begin{cases}
p_1^{(n)}(\infty)=\left[p_1^{(n-1)}(\infty)\mathrm{e}^{v}+\left(1-p_1^{(n-1)}(\infty)\right)\mathrm{e}^{-v}\right]Q_{n-1}/Q_n, & n\geq 2;\\
p_i^{(n)}(\infty)=p_{i-1}^{(n-1)}(\infty)\left(\mathrm{e}^{v}+\mathrm{e}^{-v}\right)Q_{n-1}/Q_n, n\geq 2, & i=2,\ldots,n.
\end{cases}
\tag{7}
$$

Equation (7) readily gives $p_i^{(n)}(\infty)=1/2$, and in fact the above results can easily be obtained from the symmetry of the straight strings. The main purpose of using the methodology described above to reach Eq. (7) is to present useful mathematical methods to deduce the more complicated $p_i^{(n)}(0)$ parameter.

By taking a 4-string as an example, the calculation processes of $p_i^{(n)}(0)$ are

$$
\begin{aligned}
p_1^{(4)}(0) &= q_1^{(4)}(0)+q_2^{(4)}(0)+q_3^{(4)}(0)+q_4^{(4)}(0)+q_5^{(4)}(0)+q_6^{(4)}(0)+q_7^{(4)}(0)+q_8^{(4)}(0)\\
&= \left[\mathrm{e}^{-e_1^{(4)}(F_0)}+\mathrm{e}^{-e_2^{(4)}(F_0)}+\mathrm{e}^{-e_3^{(4)}(F_0)}+\mathrm{e}^{-e_4^{(4)}(F_0)}+\mathrm{e}^{-e_5^{(4)}(F_0)}+\mathrm{e}^{-e_6^{(4)}(F_0)}+\mathrm{e}^{-e_7^{(4)}(F_0)}+\mathrm{e}^{-e_8^{(4)}(F_0)}\right]\Big/Q_4\\
&= \left[\begin{array}{l}\mathrm{e}^{-e_1^{(3)}(F_0)+v+f}+\mathrm{e}^{-e_2^{(3)}(F_0)+v+f}+\mathrm{e}^{-e_3^{(3)}(F_0)+v+f}+\mathrm{e}^{-e_4^{(3)}(F_0)+v+f}\\ +\mathrm{e}^{-e_5^{(3)}(F_0)-v+f}+\mathrm{e}^{-e_6^{(3)}(F_0)-v+f}+\mathrm{e}^{-e_7^{(3)}(F_0)-v+f}+\mathrm{e}^{-e_8^{(3)}(F_0)-v+f}\end{array}\right]\Big/Q_4\\
&= \left[\left(q_1^{(3)}(0)+q_2^{(3)}(0)+q_3^{(3)}(0)+q_4^{(3)}(0)\right)\mathrm{e}^{v}+\left(q_5^{(2)}(0)+q_6^{(2)}(0)+q_7^{(3)}(0)+q_8^{(3)}(0)\right)\mathrm{e}^{-v}\right]\mathrm{e}^{f}Q_3\Big/Q_4\\
&\approx \left[p_1^{(3)}(0)\mathrm{e}^{v}+\left(1-p_1^{(3)}(0)\right)\mathrm{e}^{-v}\right](1+f)\,Q_3\Big/Q_4,
\end{aligned}
\tag{8}
$$

$$
\begin{aligned}
p_2^{(4)}(0) &= q_1^{(4)}(0)+q_2^{(4)}(0)+q_3^{(4)}(0)+q_4^{(4)}(0)+q_9^{(4)}(0)+q_{10}^{(4)}(0)+q_{11}^{(4)}(0)+q_{12}^{(4)}(0)\\
&= \left[\mathrm{e}^{-e_1^{(4)}(F_0)}+\mathrm{e}^{-e_2^{(4)}(F_0)}+\mathrm{e}^{-e_3^{(4)}(F_0)}+\mathrm{e}^{-e_4^{(4)}(F_0)}+\mathrm{e}^{-e_9^{(4)}(F_0)}+\mathrm{e}^{-e_{10}^{(4)}(F_0)}+\mathrm{e}^{-e_{11}^{(4)}(F_0)}+\mathrm{e}^{-e_{12}^{(4)}(F_0)}\right]\Big/Q_4\\
&= \left[\begin{array}{l}\mathrm{e}^{-e_1^{(3)}(F_0)+v+f}+\mathrm{e}^{-e_2^{(3)}(F_0)+v+f}+\mathrm{e}^{-e_3^{(3)}(F_0)+v+f}+\mathrm{e}^{-e_4^{(3)}(F_0)+v+f}\\ +\mathrm{e}^{-e_1^{(3)}(F_0)-v-f}+\mathrm{e}^{-e_2^{(3)}(F_0)-v-f}+\mathrm{e}^{-e_3^{(3)}(F_0)-v-f}+\mathrm{e}^{-e_4^{(3)}(F_0)-v-f}\end{array}\right]\Big/Q_4\\
&= \left(q_1^{(3)}(0)+q_2^{(3)}(0)+q_3^{(3)}(0)+q_4^{(3)}(0)\right)\left(\mathrm{e}^{v+f}+\mathrm{e}^{v-f}\right)Q_3\Big/Q_4\\
&\approx p_1^{(3)}(0)\,(2\mathrm{ch}v+2\mathrm{sh}v\cdot f)\,Q_3\Big/Q_4, \qquad (9)
\end{aligned}
$$

where $\mathrm{sh}v\equiv(\mathrm{e}^v-\mathrm{e}^{-v})/2$.

Both the renumbering of the orientational states shown in Table 1 and the symmetry of the straight strings show that $p_i^{(n)}(0)=p_{n-i+1}^{(n)}(0)$. Table 4 presents the following quantities calculated from $n=1$ to 13 using the method indicated above, in which, $\bar{p}_1^{(n)}(0)\equiv p_1^{(n)}(0)/\left[p_1^{(n-1)}(0)\,(1+f)\,\mathrm{th}v\right]$, and $\bar{p}_i^{(n)}(0)\equiv\left(p_i^{(n)}(0)/p_1^{(n-i)}(0)-1\right)/\mathrm{th}v\cdot f$, where $\mathrm{th}v\equiv(\mathrm{e}^v-\mathrm{e}^{-v})/(\mathrm{e}^v+\mathrm{e}^{-v})$.

**Table 4.** Calculated $\bar{p}_i^{(n)}(0)$ of the straight $n$-string for $n=1$ to 13 in the external field of Eq. (4).

| $n$ | $\bar{p}_1^{(n)}(0)$ | $\bar{p}_2^{(n)}(0)$ | $\bar{p}_3^{(n)}(0)$ | $\bar{p}_4^{(n)}(0)$ | $\bar{p}_5^{(n)}(0)$ | $\bar{p}_6^{(n)}(0)$ | $\bar{p}_7^{(n)}(0)$ |
|---|---|---|---|---|---|---|---|
| 2 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(1)}(0)2\mathrm{sh}v}$ | | | | | | |
| 3 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(2)}(0)2\mathrm{sh}v}$ | 1 | | | | | |
| 4 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(3)}(0)2\mathrm{sh}v}$ | 1 | | | | | |
| 5 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(4)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | | | | |
| 6 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(5)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | | | | |
| 7 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(6)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | | | |
| 8 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(7)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | | | |
| 9 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(8)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | $\dfrac{4\mathrm{e}^{3v}+4\mathrm{e}^{-v}}{(2\mathrm{ch}v)^3}$ | | |
| 10 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(9)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | $\dfrac{4\mathrm{e}^{3v}+4\mathrm{e}^{-v}}{(2\mathrm{ch}v)^3}$ | | |
| 11 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(10)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | $\dfrac{4\mathrm{e}^{3v}+4\mathrm{e}^{-v}}{(2\mathrm{ch}v)^3}$ | $\dfrac{5\mathrm{e}^{4v}+10+\mathrm{e}^{-4v}}{(2\mathrm{ch}v)^4}$ | |
| 12 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(11)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | $\dfrac{4\mathrm{e}^{3v}+4\mathrm{e}^{-v}}{(2\mathrm{ch}v)^3}$ | $\dfrac{5\mathrm{e}^{4v}+10+\mathrm{e}^{-4v}}{(2\mathrm{ch}v)^4}$ | |
| 13 | $1+\dfrac{\mathrm{e}^{-v}}{p_1^{(12)}(0)2\mathrm{sh}v}$ | 1 | $\dfrac{2\mathrm{e}^v}{2\mathrm{ch}v}$ | $\dfrac{3\mathrm{e}^{2v}+\mathrm{e}^{-2v}}{(2\mathrm{ch}v)^2}$ | $\dfrac{4\mathrm{e}^{3v}+4\mathrm{e}^{-v}}{(2\mathrm{ch}v)^3}$ | $\dfrac{5\mathrm{e}^{4v}+10+\mathrm{e}^{-4v}}{(2\mathrm{ch}v)^4}$ | $\dfrac{6\mathrm{e}^{5v}+20\mathrm{e}^v+6\mathrm{e}^{-3v}}{(2\mathrm{ch}v)^5}$ |

The results in Table 4 indicate: (i) each column of $\bar{p}_i^{(n)}(0)$ has the same value; (ii) from the 2nd column, $p_1^{(n)}(0)=\left[p_1^{(n-1)}(0)\mathrm{e}^v+\left(1-p_1^{(n-1)}(0)\right)\mathrm{e}^{-v}\right](1+f)/2\mathrm{ch}v$; (iii) for the 3rd to 8th columns, the values of $n$, corresponding to the first appearing terms of the $\bar{p}_{i+1}^{(n)}(0)$ from top to bottom, are always odd and $n=2i+1$, so all $\bar{p}_j^{(n)}(0)$ can be calculated if the general form of $\bar{p}_{i+1}^{(2i+1)}(0)$ is found; (iv) the denominator of $\bar{p}_j^{(n)}(0)$ from $j$=2 to 7 is $(2\mathrm{ch}v)^{j-1}$, but the numerator is more complex though the following regularities can be found: (i) the coefficients of the exponential are parts of the combinatory number $C_i^l$, $l=0,\ldots,i$, the multiplying

exponential is $e^{(i-1)v}$, and each exponential decreases by a factor $e^{4v}$, e.g., $e^{(i-5)v}$, $e^{(i-9)v}$, $e^{(i-13)v}$; (ii) since each exponential factor of the $i$th ordinal in $(2\mathrm{ch}v)^i = \sum_{l=0}^{i} C_i^l e^{(i-2l)v}$, decreases by a factor $e^{2v}$, it can be found that the coefficients of the exponential terms of the numerator of $p_{i+1}^{(2i+1)}(0)$ are $C_i^{2l+1}, l = 1, \ldots,$ int $[(i-1)/2]$, where int $[(i-1)/2]$ is an integer $x$. Then, the relationship

$$p_{i+1}^{(2i+1)}(0) = p_1^{(i+1)}(0)\left[1 + \frac{\mathrm{th}v}{(2\mathrm{ch}v)^{i-1}} \sum_{l=0}^{\mathrm{int}[(i-1)2]} C_i^{2l+1} e^{(i-1-4l)v} f\right], \ i \geq 1$$

can be obtained. In view of this, the recursive equation of $p_i^{(n)}(0)$ for arbitrary n is obtained from Table 4 as follows:

$$\begin{cases} p_1^{(1)}(0) = \dfrac{1-f}{2}, \\ p_1^{(n)}(0) = \left[p_1^{(n-1)}(0)e^v + \left(1 - p_1^{(n-1)}(0)\right)e^{-v}\right]\dfrac{1+f}{2\mathrm{ch}v}, & n \geq 2, \\ p_{i+1}^{(2i+m)}(0) = p_1^{(i+m)}(0)\left[1 + \dfrac{\mathrm{th}v}{(2\mathrm{ch}v)^{i-1}} \displaystyle\sum_{l=0}^{\mathrm{int}[(i-1)/2]} C_i^{2l+1} e^{(i-1-4l)v} f\right], & i \geq 1, m \geq 1. \end{cases} \tag{10}$$

Moreover, the simplified form of Eq. (10) is

$$\begin{cases} p_1^{(n)}(0) = \dfrac{1}{2}\left[1 + \displaystyle\sum_{l=0}^{n-1} (\mathrm{th}v)^l f\right], & n \geq 1; \\ p_{i+1}^{(2i+m)}(0) = \dfrac{1}{2}\left\{1 + \left[\displaystyle\sum_{l=0}^{i+m-1} (\mathrm{th}v)^l + \frac{\mathrm{th}v}{(2\mathrm{ch}v)^{i-1}} \sum_{l=0}^{\mathrm{int}[(i-1)/2]} C_i^{2l+1} e^{(i-1-4l)v}\right] f\right\}, & i \geq 1, m \geq 1. \end{cases} \tag{11}$$

Finally,

$$\begin{cases} \delta_1^{(n)}(0) = \dfrac{1}{2}\displaystyle\sum_{l=0}^{n-1} (\mathrm{th}v)^l f, & n \geq 1; \\ \delta_{i+1}^{(2i+m)}(0) = \dfrac{1}{2}\left[\displaystyle\sum_{l=0}^{i+m-1} (\mathrm{th}v)^l + \frac{\mathrm{th}v}{(2\mathrm{ch}v)^{i-1}} \sum_{l=0}^{\mathrm{int}[(i-1)/2]} C_i^{2l+1} e^{(i-1-4l)v}\right] f, & i \geq 1, m \geq 1. \end{cases} \tag{12}$$

Equation (12) easily leads to two important conclusions: (i) $\delta_i^{(n)}(0) = f/2$ for $v = 0$, the case of an independent molecule Debye relaxation in the double-wells,[20] and (ii) $\delta_i^{(n)}(0) = nf/2$ for $v \to \infty$, reflecting the case of the strong correlation limit where the n molecules of an $n$-string behave like a single one. For arbitrary values of $v \equiv V/k_\mathrm{B}T$ and $n$, the values calculated for $\delta_i^{(n)}(0)$ are shown in Figs. 1 to 3.

Figure 1 indicates that $\delta_i^{(101)}(0)$ increases with increasing $v$ as a whole. The following points should be highlighted: (i) for small values of $i$ ($i < 10$), $\delta_i^{(101)}(0)$ goes up rapidly when $v = 1$, but it is almost constant for large values of $i$ ($10 < i \leq 51$); (ii) $\delta_i^{(101)}(0)$ keeps a slow tendency to increase with increasing $i$ for $v = 2$; and (iii) the value of $\delta_i^{(101)}(0)$ is nearly independent of $i$ when $v = 5$.

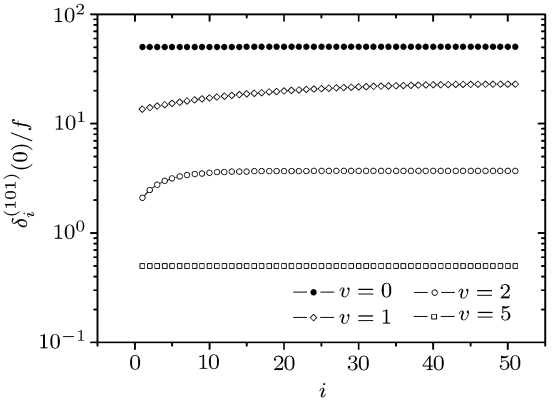



**Fig. 1.** The $\delta_i^{(101)}(0)/f$ as a function of $i$ for a set of positive $v$ values.

To make evident the changes of $\delta_i^{(n)}(0)$ with $v$, values of $\delta_i^{(n)}(0)$ are plotted as a function of $v$ in Fig. 2 for $n = 3$, 11, 21, 41, 61, 81 and 101. The results indicate that $\delta_i^{(n)}(0)$ is relatively small for $v < 1$, but it goes up rapidly as $v$ increases from 2 to 4. However, $\delta_i^{(n)}(0)$ becomes independent of $v$for $v > 5$.

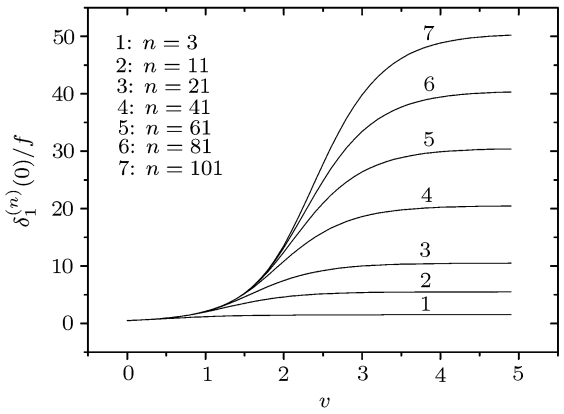



**Fig. 2.** The $\delta_i^{(n)}(0)/f$ as a function of $v$ for a set of $n$ values.

The parameter $\delta_i^{(101)}(0)$ was calculated for negative values of $v$, specifically $v = -0.5$, $-1$, $-2$, and $-4$, and the pertinent plots $\delta_i^{(101)}(0)$ vs $v$ are shown in Fig. 3. The most important difference from the results obtained for $\nu > 0$ is that $\delta_i^{(n)}(0)$ shows a periodic decrease with $n$, the period being 2 for $v < 0$. Specifically, the vibration region increases but the equilibrium value decreases with increasing $|v|$, and the region extends to the whole string though the equilibrium value becomes almost zero.

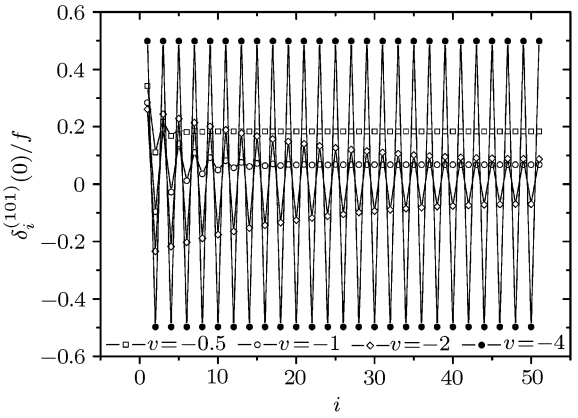



**Fig. 3.** Variation of $\delta_i^{(101)}(0)/f$ as a function of $i$ for a set of negative $v$ values.

Based on the calculated SIC, the general initial condition of the SRE can be calculated (to be published). By comparing experiments, we can check whether the model gives a good and unified account of $\alpha$- and Johari–Goldstein $\beta$-relaxation on molecular level. These items will be described in separate publications.

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