Dynamical Form-Factor of Neutron Scattering by DNA Solitons

L.V. YAKUSHEVICH

*Institute of Cell Biophysics of the Russian Academy of Sciences, Pushchino, Moscow region
142292, Russia*

Abstract. Method of scattering of slow neutrons has been considered in recent years as a rather perspective way to study the nonlinear DNA dynamics. In this paper we present the results of theoretical calculations of the dynamical form-factor of the scattering. The calculations were made on the basis of the nonlinear mathematical model which takes into account rotational motions of bases around the sugar-phosphate chains. The results of the calculations are considered as predictions for further neutron scattering experiments.

Key words: DNA, neutron scattering, nonlinear dynamics, soliton

Introduction

It is now widely accepted that internal dynamics of the DNA molecule and especially internal motions of large amplitudes, such as opening of base pairs or local transitions between different (A-, B- or other) forms, play an important role in the functioning of DNA. In recent years these motions have been studied intensively, both theoretically and experimentally. As a result, many nonlinear mathematical models of the internal DNA dynamics have been proposed and some of them were successfully applied to explain the available experimental data [1, 2]. These explanations, however, are only a few of the possible interpretations of the experimental data. To prove the preference of interpretations based on the nonlinear concept, a more detailed experimental study of the internal DNA dynamics is required. And one of the most perspective ways of experimental studying of the nonlinear internal DNA dynamics is the method of slow neutron scattering.

A possibility of using the method of slow neutron scattering to study the internal DNA dynamics was discussed intensively at the International Workshop 'Nonlinear excitations in Biomolecules' (Les Houches, May 30 – June 4, 1994) [3, 4]. From this discussion it followed that for realization of the program on neutron scattering by DNA it was necessary 1) to have a possibility to prepare a sample of oriented DNA molecules with the size appropriate for neutron experiment, 2) to find the equipment for neutron scattering experiment, and 3) to make preliminary

calculations of the spectrum of neutron scattering, which could give a possibility to organize neutron scattering experiments in a more effective way.

The first part of the problem has been solved already and an appropriate sample has been demonstrated by Grimm at the same Workshop [3]. It also seems that there is a possibility to solve the second part by using basic facilities of Scientific Nuclear Centers, for example the facilities of the Joint Institute for Nuclear Research (JINR) in Dubna (Russia) where radiobiological investigation is one of the main lines of the program of the scientific research plan [5]. One of the solutions of the third part of the problem is the theme of this paper. We present here the results of theoretical calculations of the dynamical form-factor of slow neutron scattering by DNA. The calculations were made by the technique described in [6] with the using of nonlinear mathematical model proposed in [7].

Method of Calculation

Neutron spectrometers usually provide data in the form of a double differential cross-section, $\partial^2\sigma/d\Omega dE'$. So, the aim of these theoretical calculations is also to obtain this characteristics. The general theory [6] gives the formula for double differential cross-section in the following form

$$\partial^2\sigma/\partial\Omega dE' \sim N(k''/k')[\sigma_{inc}S_{inc}(\mathbf{x}, w) + \sigma_{coh}S_{coh}(\mathbf{x}, w)]; \quad (1)$$

where σ_{inc} and σ_{coh} are incoherent and coherent scattering cross-sections, respectively; $S_{coh}(\mathbf{x}, w)$, $S_{inc}(\mathbf{x}, w)$ are the dynamical factors of coherent and incoherent scattering. $\hbar\mathbf{x}$ and $\hbar w$ are the momentum and energy transfers, respectively ($\hbar = \text{Planck's constant}/2\pi$; $\hbar\mathbf{x} = \hbar(\mathbf{k}' - \mathbf{k}'')$; $\hbar w = E' - E''$; the initial and final wave vectors and energies are denoted by $\mathbf{k}'$, $\mathbf{k}''$ and E' , E''). In the nonlinear dynamical models of DNA we shall use for calculations, the DNA bases play the role of target nuclei.

The fraction of radiation which is scattered inelastically, is of most interest because it is determined by the exchange of energy between neutrons and nonlinear excitations in the sample. Just this inelastic scattering carries information about internal motions in the system investigated. According to the general theory [6] the formula for the dynamical factor of coherent inelastic scattering is

$$S_{coh.,inel.}(\mathbf{x}, w) = [\exp(-2W_{\mathbf{x}})/(4\pi\hbar N)]\Sigma_n \Sigma_n, \Sigma_j \Sigma_j, \exp[-i\mathbf{x}(\mathbf{R}_{n,j}^0 - \mathbf{R}_{n',j'}^0)] \int dt \exp(-iwt) \langle \exp(-i\mathbf{x}\mathbf{u}_{n,j}(0)), \exp(i\mathbf{x}\mathbf{u}_{n',j'}(t)) \rangle; \quad (2)$$

where $\mathbf{R}_{n,j}^0$ is the position vector of the n -th base in the j -th DNA strand ($j = 1, 2$); $\mathbf{u}_{n,j}$ is a possible displacement of the n -th base of the j -th strand from its equilibrium position; N is the general number of bases in one of the strands; $\exp(-2W_{\mathbf{x}})$ is the Debye-Waller factor.

Formula (2) contains the correlation function $\langle \exp(-i\mathbf{x}\mathbf{u}_{n,j}(0)), \exp(i\mathbf{x}\mathbf{u}_{x'j'}(t)) \rangle$ and now the problem is how to calculate it. In other words, the problem is how to calculate the correlation function for the assemblage of DNA solitons.

To solve the problem, let us use the approach proposed by Mikeska [8] and developed in one of our previous works [9]. This approach takes into account that the dynamical behavior of DNA solitons is very similar to the dynamical behavior of usual classical particles [10]. So, in the first approximation, instead of the assemblage of solitons we can consider an assemblage of usual classical particles with the energies, sizes, velocities and masses of solitons. Moreover, we can suggest that the number of the particles (solitons) is not large and that the ideal gas approximation is valid. As a result, we can use the following approximation formula

$$\langle \dots \rangle \cong \bar{N}_s \langle \dots \rangle_1; \quad (3)$$

where $\bar{N}_s$ is the average number of solitons, and $\langle \dots \rangle_1$ denotes the averaging over the states of an individual soliton. This simplification gives us a possibility to calculate without any problems the dynamical factor of inelastic coherent scattering (2). The only thing which still remains indefinite is the model of DNA internal dynamics.

The Model

Let us model DNA by two parallel chains of pendula interacting with each other by longitudinal and transverse springs (Figure 1). Here pendula imitate DNA bases, two chains of longitudinal springs imitate sugar-phosphate chains and transverse springs imitate hydrogen bonds between bases. This model can be considered as a modification of the well-known mechanical model proposed by Scott [11] and applied to DNA by Englander et al. [12] (Figure 2). The model consists of one horizontal chain of pendula placed in the gravitational field of the Earth, each pendulum being able rotate in the xy plane. To receive a modified model it is enough to 'remove' the gravitational field and to 'add' one more chain of pendula and transverse springs imitating weak interactions between pendula of different chains. The resulting model shown in Figure 1 looks like a one-dimensional lattice with two pendula per unit cell. So, to describe the model system, we can introduce the basic vector $\mathbf{a} = \{0; 0; a\}$, and the lattice vector $\mathbf{R}_n^0$ defined as $\mathbf{R}_n^0 = n\mathbf{a}$, where a is the distance between the neighboring pendula and n is the integer ($n = 1, 2, \dots, N$).

The equilibrium positions of the masses of pendula within a unit cell can be denoted by vectors $\mathbf{d}_1 = \{l; 0; 0\}$ and $\mathbf{d}_2 = \{b - l; 0; 0\}$, where b is the distance between the chains and l is the length of pendula. Then the position vector $\mathbf{R}_{n,j}^0$ of the j -th pendulum mass ($j = 1, 2$) in the n -th cell is given by

$$\mathbf{R}_{n,j}^0 = \mathbf{R}_n^0 + \mathbf{d}_j. \quad (4)$$

Taking into account that every pendulum rotates only in the xy plane, we can write possible displacements of masses as

Figure 1. Double chain of pendula (a), and possible rotational displacements $(\varphi_{n,1}, \varphi_{n,2})$ of the n -th pendula in the xy plain (b).

Figure 2. Chain of pendula.

$$\begin{aligned} \mathbf{u}_{n,1} &= \mathbf{R}_{n,1}(t) - \mathbf{R}_{n,1}^0 = \{-l(1 - \cos \varphi_{n,1}); l \sin \varphi_{n,1}; 0\}; \\ \mathbf{u}_{n,2} &= \mathbf{R}_{n,2}(t) - \mathbf{R}_{n,2}^0 = \{l(1 - \cos \varphi_{n,2}); l \sin \varphi_{n,2}; 0\}; \end{aligned} \quad (5)$$

where $\varphi_{n,j}$ is the angular displacement of the n -th pendulum of the j -th chain.

The model Hamiltonian will have then the form

$$\begin{aligned} H &= \sum_n \{M \dot{\mathbf{u}}_{n,1}^2/2 + M \dot{\mathbf{u}}_{n,2}^2/2\} + \sum_n \{K |\mathbf{u}_{n,1} - \mathbf{u}_{n-1,1}|^2/2 \\ &\quad + K |\mathbf{u}_{n,2} - \mathbf{u}_{n-1,2}|^2/2 + k |\mathbf{u}_{n,1} - \mathbf{u}_{n,2}|^2/2\} \end{aligned} \quad (6)$$

where K and k are the rigidities of longitudinal and transverse springs, correspondingly. If we insert now (5) into (6) and use 1) linear approximation according to φ in the terms describing interactions along the chains (rigid springs) and 2) nonlinear approximation in the terms describing interactions between chains (soft springs), then the final model Hamiltonian takes the form

$$H = \sum_n \{ M \dot{\varphi}_{n,1}^2 / 2 + M \dot{\varphi}_{n,2}^2 / 2 \} + \sum_n \sum_j K l^2 \{ |\varphi_{n,j} - \varphi_{n-1,j}|^2 \} / 2 + \\ + \sum_n k l^2 \{ 2(1 - \cos \varphi_{n,1}) + 2(1 - \cos \varphi_{n,2}) - [1 - \cos(\varphi_{n,1} + \varphi_{n,2})] \}. \quad (7)$$

Corresponding dynamical equations are

$$I \ddot{\varphi}_{n,1} = K l^2 (\varphi_{n+1,1} - 2\varphi_{n,1} + \varphi_{n-1,1}) - k l^2 [2 \sin \varphi_{n,1} - \sin(\varphi_{n,1} + \varphi_{n,2})]; \\ I \ddot{\varphi}_{n,2} = K l^2 (\varphi_{n+1,2} - 2\varphi_{n,2} + \varphi_{n-1,2}) - k l^2 [2 \sin \varphi_{n,2} - \sin(\varphi_{n,2} + \varphi_{n,1})]; \quad (8)$$

where $I = M l^2$.

In the continuum limit when

$$a \rightarrow 0; \quad N \rightarrow \infty; \quad n a \rightarrow z; \quad \varphi_{n,j}(t) \rightarrow \varphi_j(z, t); \quad \Sigma \rightarrow \int dz;$$

Equations (8) can be rewritten in the form

$$I \varphi_{1tt} = K l^2 a^2 \varphi_{1zz} - k l^2 [2 \sin \varphi_1 - \sin(\varphi_1 + \varphi_2)]; \\ I \varphi_{2tt} = K l^2 a^2 \varphi_{2zz} - k l^2 [2 \sin \varphi_2 - \sin(\varphi_2 + \varphi_1)]. \quad (9)$$

It is convenient to rewrite Equations (9) in the following way

$$\varphi_{1tt} - C_0^2 \varphi_{1zz} + w_0^2 \{ 2 \sin \varphi_1 - \sin(\varphi_1 + \varphi_2) \} = 0; \\ \varphi_{2tt} - C_0^2 \varphi_{2zz} + w_0^2 \{ 2 \sin \varphi_2 - \sin(\varphi_2 + \varphi_1) \} = 0; \quad (10)$$

where $C_0^2 = K l^2 a^2 / I$; $w_0^2 = k l^2 / I$.

The general solution of Equations (10) has not been found yet. For calculations of the dynamical factor of neutron scattering we shall use the particular solution

$$\varphi_1(z, t) = -\varphi_2(z, t) = 4 \arctan \{ \exp[\gamma(z - vt)/d] \}; \quad (\gamma = (1 - v^2/C_0^2)^{-1/2}); \quad (11)$$

which has been found by the direct algebraic method in [13]. This solution is shown schematically in Figure 3. In Figure 4 a qualitative picture corresponding to the solution is shown. Sugar-phosphate chains of DNA are shown here as long lines and the bases are shown with the help of short transverse rods. The solution has a simple interpretation of a local conformational deformation having the form of local open state of size $d = C_0/(2^{1/2}w_0)$ and moving along the DNA molecule with velocity v . This is why this solution is interesting for biological applications.

Figure 3. Two coupled kinks.

Figure 4. Moving local deformation.

Similar Models

In the model described above, the rotational motions of bases around the sugar-phosphate chains were considered. But recently a new suggestion has been put forward by Maria Barbi et al. [14]. When constructing the twist-stretched model describing the process of local unwinding of the double helix, these authors noticed that rotational motions of nucleotides around the main DNA axis (as well as the transverse motions considered earlier by Peyrard and Bishop [15]) might be also important. Let us consider independently this type of rotational motions. In this case, the vectors of displacements, $\mathbf{u}_{n,1}$, $\mathbf{u}_{n,2}$, have the form

$$\mathbf{u}_{n,1} = \mathbf{R}_{n,1}(t) - \mathbf{R}_{n,1}^0 = \{R(1 - \cos \theta_{n,1}); -R \sin \theta_{n,1}; 0\}; \quad (12)$$

$$\mathbf{u}_{n,2} = \mathbf{R}_{n,2}(t) - \mathbf{R}_{n,2}^0 = \{-R(1 - \cos \theta_{n,2}); R \sin \theta_{n,2}; 0\};$$

where $\theta_{n,i}$ is the angular displacement of the n -th nucleotide of the i -th strand around the main DNA axis (Figure 5).

If we insert (12) into the Hamiltonian (6), we obtain

$$H = \sum_n R l^2 (\dot{\theta}_{n,1}^2 + \dot{\theta}_{n,2}^2) / 2 + \sum_n K R^2 [1 - \cos(\theta_{n,1} - \theta_{n-1,1})]$$

Figure 5. A schematic picture of the double polynucleotide chain (a); possible displacements ($\mathbf{u}_{n,1}$, $\mathbf{u}_{n,2}$) of the nucleotides into the xy plane (b), $\theta_{n,1}$, $\theta_{n,2}$ being the angles of rotations.

$$\begin{aligned}
 &+KR^2[1 - \cos(\theta_{n,2} - \theta_{n-1,2})] + kR^2[2(1 - \cos \theta_{n,1}) + 2(1 - \cos \theta_{n,2}) \\
 &+(1 - \cos(\theta_{n,1} - \theta_{n,2}))].
 \end{aligned} \tag{13}$$

From mathematical point of view, Hamiltonian (13) is very similar to Hamiltonian (7) and to the Hamiltonians proposed earlier by Takeno and Homma [16], Yomosa [17], Fedyanin and co-authors [18], and Zhang [19]. So, we can expect that the results of calculations of the dynamical factor made for the model (7) and presented in the next section, should be valid for all similar models mentioned above.

Results of Calculations of the Dynamical Form-Factor and Discussion

The algorithm of calculation of the dynamical factor includes three steps. We need

- (i) to insert (5) and (11) into (2);
- (ii) to pass to the continuum approximation ($a \rightarrow 0$; $N \rightarrow \infty$; $na \rightarrow z$; $\varphi_{n,j}(t) \rightarrow \varphi_j(z, t)$; $\Sigma \rightarrow \int dz$) and
- (iii) to apply approximation of the ideal gas.

As a result, we obtain

$$S_{coh.,inel.}(\mathbf{x}, w) = \{[4l^2\gamma_0 d \exp(-2W_{\mathbf{x}})(E_0/2\pi k_B T)^{1/2}]/[\hbar C_0 a x_z k_1(E_0/k_B T)]\} \\ \exp(-E_0/k_B T) \exp(-E_0\gamma_0/k_B T) \{X^2(1 + \cos x_y h) - 2XY \sin x_y h + \\ + Y^2(1 - \cos x_y h)\}; \quad (14)$$

where $X = x_x(\pi dx_z/\gamma_0)/sh(\pi dx_z/2\gamma_0)$; $Y = x_y(\pi dx_z/\gamma_0)/ch(\pi dx_z/2\gamma_0)$; $\gamma_0 = (1 - v_0^2/C_0^2)^{-1/2}$; $v_0 = w/x_z$; $\mathbf{x} = \{x_x; x_y; x_z\}$; $E_0 = 8l^2(2Kk)^{1/2}$; $h = b - 2l$; $k_1(z)$ is the Macdonald function [20]

$$k_v(z) = \int_0^\infty \exp[-zch(t)]ch(vt)dt. \quad (15)$$

Formula (14) shows that in the spectrum of scattering there is a central peak with characteristics depending on the wave vector $\mathbf{x}$ and on temperature T . The width, Δw , and integral intensity, I , of the peak can be easily estimated in the same way as it was done in [9]. It can be shown, for example, that the dependence of I and Δw on temperature is

$$I = \int S_{coh.,inel.}(\mathbf{x}, w)dw = A(\mathbf{x})k_B T \exp(-E/k_B T); \quad (16) \\ \Delta w = B(\mathbf{x})(k_B T)^{1/2};$$

where A and B are temperature-independent coefficients.

Formula (14) can be improved by taking into account the helical structure of DNA. One can do it in two different ways. The first way is based on the method proposed in [9], and we can expect that application of this method will give a possibility to obtain the splitting of the central peak ($x_z \Rightarrow x_z \pm 2\pi/10a$) due to the helical structure of DNA. The second way is based on the approach proposed by Gaeta [21, 22]. In this approach additional interactions due to the helical structure are taken into account, and this in turn leads to renormalization of the parameters of the model hamiltonian. As a result, we can expect to obtain at least the shift of the central peak.

Acknowledgment

This work was supported by the Russian Foundation for Basic Research (Grant N97-04-48417).

References

1. Yakushevich, L.V.: Nonlinear dynamics of biopolymers: theoretical models, experimental data, *Quart. Rev. Biophys.* **26** (1993), 201–223.

2. Yakushevich, L.V.: *Nonlinear physics of DNA*, Wiley and Sons, Chichester, New York, Brisbane, Toronto, Singapore, 1998.
3. Grimm, H. and Rupprecht, A.: Inelastic neutron scattering studies of oriented DNA. In M. Peyrard (ed.), *Nonlinear excitations in biomolecules*, Springer-Verlag, Berlin, Heidelberg, New York, 1995, pp. 101–115.
4. Middendorf, H.D.: Neutron scattering studies of biopolymer-water systems: solvent mobility and collective excitations. In M. Peyrard (ed.), *Nonlinear excitations in biomolecules*, Springer-Verlag, Berlin, Heidelberg, New York, 1995, pp. 369–383.
5. Aleinikov, V.E. and Krasavin, E.A.: Research program of the division of radiation and radiobiological research: its transformance in 1997 and the program for 1998. Report to the 83rd Session of the Scientific Council of JINR, January 15–16, 1998, *JINR-97-375*, Dubna, 1997.
6. Marshall, W. and Lovasey, S.W.: *Theory of thermal neutron scattering*, Oxford University Press, Oxford, 1971, Chapter 4.3.
7. Yakushevich, L.V.: Nonlinear DNA dynamics: a new model, *Phys. Letters A*-**136** (1989), 413–417.
8. Mikeska, H.J.: Solitons in a one-dimensional magnet with an easy plane, *J. Phys. C: Solid State Phys.* **11** (1978), L29–L32.
9. Fedyanin, V.K. and Yakushevich, L.V.: Scattering of neutrons and light by DNA solitons, *Studia. biophys.* **103** (1984), 171–178.
10. Yakushevich, L.V.: The effect of damping, external fields and inhomogeneity on the nonlinear dynamics of biopolymers, *Studia biophys.* **121** (1987), 201–207.
11. Scott, A.C.: A nonlinear Klein-Gordon equation, *Am. J. Phys.* **37** (1969), 52–61.
12. Englander, S.W., Kallenbach, N.R., Heeger, A.J., Krumhansl, J.A. and Litwin, A.: Nature of the open state in long polynucleotide double helices: possibility of soliton excitations, *Proc. Natl. Acad. Sci. U.S.A.* **77** (1980), 7222–7226.
13. Yakushevich, L.V.: An exact solution to the system of nonlinear differential equations simulating torsional dynamics of DNA, *Russian Journal of Physical Chemistry* **69** (1995), 1277–1280.
14. Barbi, M., Cocco, S., Peyrard, M. and Ruffo, S.: A twist-stretch model of DNA, *Abstracts of the International conference 'Nonlinear Phenomena in Biology, June 23–June 28, 1998'*, Institute of Cell Biophysics of the Russian Academy of Sciences, Pushchino, Russia, p. 5.
15. Peyrard, M. and Bishop, A.R. Statistical mechanics of a nonlinear model for DNA denaturation, *Phys. Rev. Letters* **62** (1989), 2755–2758.
16. Takeno, S. and Homma, S.: Topological solitons and modulated structure of bases in DNA double helices, *Prog. Theor. Phys.* **70** (1983), 308–311.
17. Yomosa, S.: Soliton excitations in deoxyribonucleic acid (DNA) double helices, *Phys. Rev. A*-**27** (1983), 2120–2125.
18. Fedyanin, V.K. and Lisy, V.: Soliton conformational excitations in DNA, *Stud. biophys.* **116** (1986), 65–71.
19. Zhang, Ch.-T.: Soliton excitations in deoxyribonucleic acid (DNA) double helices, *Phys. Rev. A*-**35** (1987), 886–891.
20. Gradstein, I.S. and Ryzhik, I.M.: *Tables of integrals, sums, series and products*, Nauka, Moscow, 1971, Chapter 8.4.
21. Gaeta, G.: On the model of DNA torsion dynamic, *Phys. Letters A*-**143** (1990), 227–232.
22. Gaeta, G.: Solitons in planar and helicoidal Yakushevich model of DNA dynamics, *Phys. Letters A*-**168** (1992), 383–389.