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ON CONFORMATIONS OF THE SUPERHELIX STRUCTURE

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The slight deformation of a helical macromolecule leading to the superhelical structure is considered. General equations which connect "internal" stereochemical parameters of the backbone of a helical macromolecule with "external" parameters of the superhelix are obtained; they are analogous to those of Shimanouchi and Mizushima. The case when the radius of the major helix is much greater than the radius of the minor helix is treated. Assuming that all conformational changes are due to small distortions of the rotation angles (bond angles and bond lengths are kept constant) the general equations reduce to a set of nonhomogeneous linear algebraic equations. Its solution (in the case of the DNA double-helix in B-form) shows that the DNA backbone can form a coiled-coil with parameters close to those estimated from experimental data on DNP in chromatine from nuclei of cells.

1. Introduction

In a number of experimental investigations the structure of chromatine has been studied. It was found that in DNP (i.e. in complexes with histones) the molecule of DNA, being mainly in the B-configuration, is folded in a regular compact structure with identity period 120 Å [1–4]. In a model recently proposed [5] the structure of chromatine is based on a repeat unit containing histones and a coiled segment of DNA (about 200 base pairs). Such units form a flexibly jointed chain of a chromatine fiber. The question arises how the DNA segment is coiled within the unit.

Periodic kinks of the DNA molecule resulting in coiling have been discussed recently [6]. In this paper we concern a possible "smooth" coiling of the DNA molecule. First of all we have to find general equations to evaluate the changes of the stereochemical parameters of helical macromolecules for which the axis of the helix has been twisted into a helix with large radius (i.e., forming superhelix).

2. Model

We assume that a helical macromolecule of arbitrary structure, but constant helix-parameters (the radius of helix ρ, the angle of rotation about the helix axis ϑ and the translation d along the helix axis per repeat unit of the chain), is coiled into a major helix of radius $R > \rho$, angle of rotation χ and translation H along the major helix axis per just the same structural unit of the chain. The coordinates of the structural units must satisfy the discrete parametric equation of a superhelix [7] in a fixed coordinate system (X, Y, Z) (see fig. 1)

$$\begin{aligned} X_k &= [R + \rho\cos(k\vartheta)]\cos(k\chi) \\ &\quad + \rho\cos\psi\sin(k\chi)\sin(k\vartheta), \\ Y_k &= -[R + \rho\cos(k\vartheta)]\sin(k\chi) \\ &\quad + \rho\cos\psi\cos(k\chi)\sin(k\vartheta), \\ Z_k &= kH + \rho\sin\psi\sin(k\vartheta), \end{aligned} \tag{1}$$

where ψ is the angle between the tangent to the axis

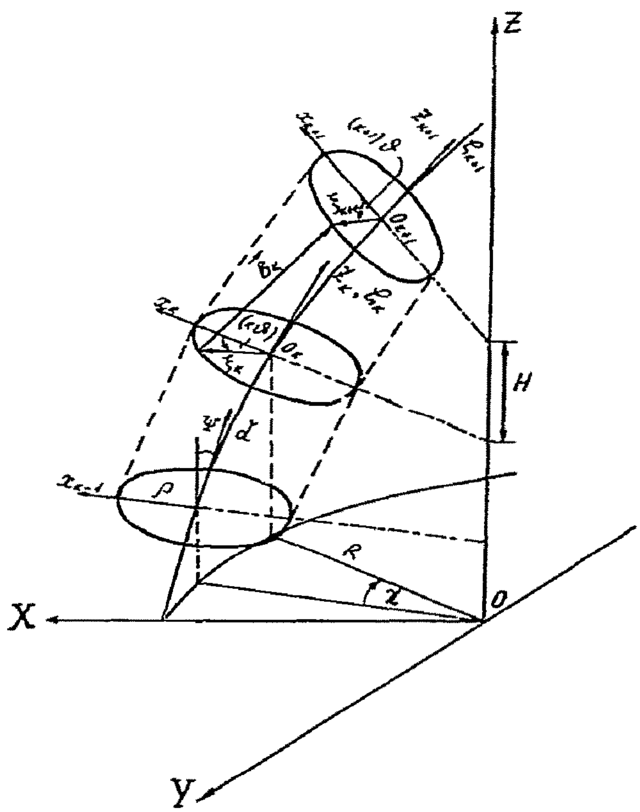

Fig. 1. Coordinate systems and parameters characterizing the major and minor helices.

of the major helix and the OZ axis and k is the number of the structural unit.

We shall use the following definitions (see also table 1): the minor helix is a helical macromolecule distorted by coiling into superhelix; the major helix is the axis of a helical macromolecule coiled into regular helix; the minor structural unit is a distorted structural unit of the ordinary helix. The nature and the extent of the distortion depend on the number of the structural unit k and vary periodically. Therefore, the term "minor structural unit" is conventional. The major structural unit is a real structural unit formed by n minor structural units. $|Oo_{k+1} - Oo_k|$ is a chord of arc $\tilde{d}$, where $\tilde{d}$ is a deformed translation along the axis of the minor helix; n is the number of the minor structural units and m is the number of turns in the identity period of the minor helix; N is the number of the major structural units and $M = (D/2\pi R)\mathrm{ctg}(\pi/2-\psi)$ is the number of turns in the identity period $D = HNn$ of the major helix.

3. Derivation of the general equations

We wish to connect the geometric characteristics of the major and minor helices with the molecular parameters of the polymer chain backbone (bond lengths l_i, bond angles θ_i and rotation angles φ_i). To this end we start from the general scheme for treating the ordinary helical structures used elsewhere [8].

Introduce three sets of cartesian coordinates (of the same sense) for each minor structural unit (see figs. 1 and 2):

Table 1
Description of helical structures

Helical structures		Characteristics of helical structures		
		repeat unit	parameters per repeat structural unit	periodicity conditions
ordinary helix of macromolecule		structural unit of p bonds	ρ d ϑ	$n\vartheta = 2\pi m$
superhelix	the minor helix	minor structural unit of p bonds	ρ $\|Oo_{k+1} - Oo_k\|$ ϑ	$n\vartheta = 2\pi m$
	the major helix	major structural unit of pn bonds	R Hn χn	$N(n\chi) = 2\pi M$

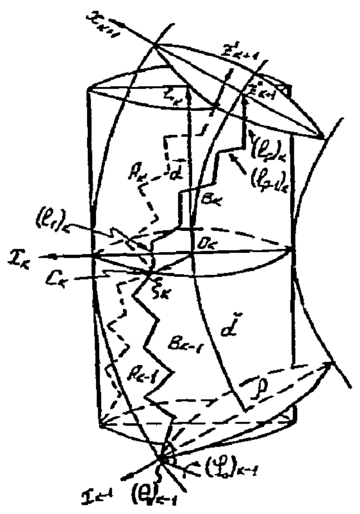

Fig. 2. Local molecular coordinate systems for distorted, (x'', y'', z''), and undistorted, (x', y', z'), sequences of structural units B_{k-1}, B_k and A_{k-1}, A_k.

$(x'', y'', z'')_k$ are local molecular coordinate systems connected with the minor structural units. The origin of each reference frame is affixed to the first atom of the structural unit.

$(x, y, z)_k$ are auxiliary local coordinate systems of the superhelix. The origin of each reference frame lies on the minor helix axis at the intersection with the perpendicular from the origin of the corresponding local molecular coordinate system. The axis z_k is directed along the tangent to the major helix in point O_k. The axis x_k is taken along the radius of the major helix.

$(\xi, \eta, \zeta)_k$ are local coordinate systems of the superhelix. They are obtained by the rotation of the coordinate system $(x, y, z)_k$ around axis z_k through the angle $(k\vartheta)$.

For any vector in the reference frame $(\xi, \eta, \zeta)_{k+1}$ transformation into its representation in the reference frame $(\xi, \eta, \zeta)_k$ through the coordinate systems of the superhelix $(x, y, z)_{k+1}$, (X, Y, Z) and $(x, y, z)_k$, on the one hand, and through the local molecular systems $(x'', y'', z'')_{k+1}$ and $(x'', y'', z'')_k$, on the other hand, yields equations which are analogous to those derived by Shimanouchi and Mizushima [8], but generalized for the superhelix. We thus obtain:

$$\hat{N}_k = \hat{T}_k^{-1} \hat{S}_k \hat{T}_{k+1}, \quad (2)$$

$$\hat{F}_k(Oo_{k+1} - Oo_k) - (\hat{E} - \hat{N}_k)\rho = \hat{T}_k^{-1} b_k. \quad (3)$$

Here $\hat{E}$ is the identity matrix, $\hat{N}_k$, $\hat{S}_k$, $\hat{F}_k$ and $\hat{T}_k$ are the transformation matrices defined by

$$\hat{N}_k : (\xi, \eta, \zeta)_{k+1} \to (\xi, \eta, \zeta)_k,$$

$$\hat{S}_k : (x'', y'', z'')_{k+1} \to (x'', y'', z'')_k,$$

$$\hat{F}_k : (X, Y, Z) \to (\xi, \eta, \zeta)_k,$$

$$\hat{T}_k : (\xi, \eta, \zeta)_k \to (x'', y'', z'')_k.$$

b_k is the "length" vector of the kth minor structural unit expressed in the local molecular coordinate system $(x'', y'', z'')_k$. As distinct from a regular cylindrical structure (ordinary helix), in our model $|b_k|$ vary periodically ($|b_k| = |b_{k+n}|$) depending on the number k of the minor structural unit. For the major structural unit the condition $|\Sigma_{i=0}^{n-1} b_i| = \text{const}$ must hold.

The elements of the matrices $\hat{N}_k$, $\hat{F}_k$ and the vector $(Oo_{k+1} - Oo_k)$ can be found in appendix 1. The matrices $\hat{S}_k$, $\hat{T}_k$ and the vector b_k depend on the choice of the local molecular coordinate systems $(x'', y'', z'')_k$. In order to find their elements, the following scheme may be used. Imagine that two successive minor structural units of the same macromolecule meet in point C_k (see fig. 2): the sequence of structural units forming a superhelix is B_{k-1}, B_k and the sequence forming an undistorted ordinary helix is A_{k-1}, A_k. The molecular coordinate system $(x'', y'', z'')_k$ for the structural unit B_k is introduced as follows: the axis z_k'' coincides with the direction of bond p in the preceding B_{k-1}, the axis x_k'' lies in a plane of bonds p and $(p-1)$ of the same unit B_{k-1}, forming an acute angle with bond $(p-1)$. The molecular coordinate system $(x', y', z')_k$ for the structural unit A_k is introduced in the same way.

Now in order to define the transformation matrix $\hat{T}_k$, it suffices to write an arbitrary vector in the coordinate system $(\xi, \eta, \zeta)_k$ and to transform it into the reference frame $(x'', y'', z'')_k$ directly and through reference frames $(x', y', z')_k$ and $(x', y', z')_{k+1}$. This leads to the expression

$$\hat{T}_k = \hat{\tilde{S}}_k \hat{S}^{-1} \hat{T}, \quad (4)$$

where $\hat{T}$ and $\hat{S}$ are transformation matrices defined by

$$\hat{T} : (\xi, \eta, \zeta)_k \to (x', y', z')_k,$$

$$\hat{S} : (x', y', z')_{k+1} \to (x', y', z')_k.$$

This performs the transformation within the sequence of the undistorted structural units A_{k-1}, A_k and therefore coincide with similar matrices in the equation of Shimanouchi and Mizushima [8]. The transformation matrix

$$\hat{\tilde{S}}_k : (x', y', z')_{k+1} \to (x'', y'', z'')_k$$

operates within the sequence B_{k-1}, A_k (because the spatial orientation of the coordinate system $(x'', y'', z'')_k$ is determined by the direction of bonds p and $(p-1)$ of the structural unit B_{k-1}).

With such molecular coordinate systems $(x', y', z')_k$ and $(x'', y'', z'')_k$ the form of the matrices $\hat{S}_k$ and $\hat{\tilde{S}}_k$ is defined, as usually, through the use of auxiliary coordinate systems $(x_i, y_i, z_i)_k$ affixed to each bond of the structural unit ($i = 1, 2, ..., p$). Then

$$\hat{S}_k = \prod_{j=0}^{p-1} (\hat{S}_j)_k, \tag{5}$$

$$\hat{\tilde{S}}_k = \prod_{j=0}^{p-1} (\hat{\tilde{S}}_j)_k. \tag{6}$$

Here $(\hat{S}_j)_k$ and $(\hat{\tilde{S}}_j)_k$ are the transformation matrices for coordinate systems $(x''_{j+1}, y''_{j+1}, z''_{j+1})_k$ and $(x''_j, y''_j, z''_j)_k$ in the sequence B_{k-1}, B_k and for coordinate systems $(x'_{j+1}, y'_{j+1}, z'_{j+1})_k$ and $(x'_j, y'_j, z'_j)_k$ in the sequence B_{k-1}, A_k, respectively. (The reference frames $(x''_0, y''_0, z''_0)_k$ and $(x'_0, y'_0, z'_0)_k$ are nothing else than $(x''_p, y''_p, z''_p)_{k-1}$.)

Taking into account that the minor structural units in the superhelix (in the identity period of the minor helix) may be distorted in different ways, we can write the following form for the matrix $(\hat{S}_j)_k$:

$$(\hat{S}_j)_k = \begin{pmatrix} -\cos(\alpha_j)_k \cos(\varphi_j)_k & \sin(\varphi_j)_k & -\sin(\alpha_j)_k \cos(\varphi_j)_k \\ -\cos(\alpha_j)_k \sin(\varphi_j)_k & -\cos(\varphi_j)_k & -\sin(\alpha_j)_k \sin(\varphi_j)_k \\ -\sin(\alpha_j)_k & 0 & \cos(\alpha_j)_k \end{pmatrix}, \tag{7}$$

$(\alpha_j)_k \equiv \pi - (\theta_j)_k$.

For an ordinary helix we have, as usually, $(\hat{S}_j)_k \equiv \hat{S}_j$ and

$$\hat{S} = \prod_{j=0}^{p-1} \hat{S}_j. \tag{8}$$

Now vector b_k can be written as

$$b_k = \sum_{t=0}^{p-1} \left[\prod_{j=0}^{t} (\hat{S}_j)_k \right] (l_{t+1})_k, \tag{9}$$

where

$$(l_{t+1})_k = \begin{pmatrix} 0 \\ 0 \\ (l_{t+1})_k \end{pmatrix}.$$

Elements of the matrix $\hat{\tilde{S}}_k$ are expressed in terms of the known rotation angles of the undistorted structural unit A_k, with the exception of the angles related to bonds $(l_1)_k$ and $(l_2)_k$ because they are defined by the $(p-1)$th and pth bonds of the structural unit B_{k-1}, according to the foregoing convention. Then eq. (6) can be written as follows

$$\hat{\tilde{S}}_k = \prod_{j=0}^{1} (\hat{\tilde{S}}_j)_k \prod_{j=2}^{p-1} \hat{S}_j, \tag{10}$$

where matrix $(\hat{\tilde{S}}_j)_k$ ($j = 0, 1$) is analogous to the transformation matrix $(\hat{S}_j)_k$ ($j = 0, 1$) [see eq. (7)] but, in general, has other values of the distorted rotation angles $(\tilde{\varphi}_j)_k$ and bond angles $(\tilde{\theta}_j)_k$.

Finally, substituting eqs. (8) and (10) in eq. (4) we have

$$\hat{T}_k = (\hat{S}_0)_k (\hat{S}_1)_k (\hat{S}_0 \hat{S}_1)^{-1} \hat{T}. \tag{11}$$

To derive the elements of the matrix $\hat{T}$ we consider three vector equations for the components of the vectors b_k, $(b_{k-1} + b_{k+1})$ and $(b_{k-1} - b_{k+1})$ in the local molecular coordinate system related to the kth structural unit of the ordinary helix

$$\begin{aligned} &\hat{T}(\hat{N} + \hat{N}^{-1})[d - (\hat{E} - \hat{N})\rho] = (\hat{S} + \hat{S}^{-1})b, \\ &\hat{T}(\hat{N} - \hat{N}^{-1})[d - (\hat{E} - \hat{N})\rho] = (\hat{S} - \hat{S}^{-1})b, \\ &\hat{T}[d - (\hat{E} - \hat{N})\rho] = b. \end{aligned} \tag{12}$$

Solving these equations for the elements of $\hat{T}$ we have

$$\hat{T} = (T_1, T_2, T_3), \tag{13}$$

where

$$T_1 = -(\rho')^{-1}(\hat{E} - \hat{S}^{-1})b,$$

$$T_2 = (\rho' \sin\vartheta)^{-1} [(\hat{E} - \hat{S}) + (\hat{E} - \hat{S}^{-1}) \cos\vartheta] b,$$

$$T_3 = [2(1 - \cos\vartheta)(d')^2]^{-1/2} (\hat{S} + \hat{S}^{-1} - 2\hat{E}\cos\vartheta) b,$$

$$\rho' = [2(\hat{E} - \hat{S}^{-1}) b \cdot b]^{1/2} = 2\rho(1 - \cos\vartheta),$$

$$d' = [(\hat{S} + \hat{S}^{-1} - 2\hat{E}\cos\vartheta) b \cdot b]^{1/2} = [2(1 - \cos\vartheta)]^{1/2} d.$$

Hence, eqs. (2) and (3) as well as eqs. (A.4), (A.5), (A.6), (5), (7), (9), (11) and (13) are general expressions connecting "internal" molecular characteristics with "external" superhelical parameters of the molecule.

At $\chi = 0$ and $\psi = 0$ or $\pi/2$ eqs. (2) and (3) are reduced to the corresponding equations for ordinary helical chains (the axis of the helix is a straight line [8]).

4. Linear approximation

We shall treat the case where $R \gg \rho$ (this is equivalent to considering only small values of χ) and the angles of internal rotation are the only parameters. Thus

$$(l_j)_k = l_j, \quad (\theta_j)_k = \theta_j,$$
$$(\varphi_j)_k = \varphi_j + (\Delta\varphi_j)_k \quad (j = 0, 1, ..., p-1), \tag{14}$$
$$(\widetilde{\varphi}_j)_k = \varphi_j + (\Delta\widetilde{\varphi}_j)_k \quad (j = 0, 1),$$

where $(\Delta\varphi_j)_k$ and $(\Delta\widetilde{\varphi}_j)_k$ are small deviations of the angles $(\varphi_j)_k$ and $(\widetilde{\varphi}_j)_k$ from the respective values of the rotation angles φ_j of the ordinary helix in sequences of structural units B_{k-1}, B_k and B_{k-1}, A_k.

Expanding components of eqs. (2), (3), (A.4), (A.5), (A.6), (5), (7), (9) and (11) into a power series of χ, $(\Delta\varphi_j)_k$ and $(\Delta\widetilde{\varphi}_j)_k$, taking into account only linear terms of the expansion, we obtain (see appendix 2) a set of non-homogeneous algebraic equations which connects all values $(\Delta\varphi_j)_k$ and $(\Delta\widetilde{\varphi}_j)_k$. The coefficients of these equations include the parameters of the undistorted helix and other molecular quantities

$$\sum_{j=0}^{p-1} T_1 \cdot l_j^0 [(\Delta\varphi_j)_k - (\delta_{0j} + \delta_{1j})(\Delta\widetilde{\varphi}_j)_k] + \sum_{j=0}^{1} a_j(\Delta\widetilde{\varphi}_j)_{k+1} = -\chi \sin\psi \sin(k\vartheta),$$

$$\sum_{j=0}^{p-1} T_2 \cdot l_j^0 [(\Delta\varphi_j)_k - (\delta_{0j} + \delta_{1j})(\Delta\widetilde{\varphi}_j)_k] + \sum_{j=0}^{1} b_j(\Delta\widetilde{\varphi}_j)_{k+1} = -\chi \sin\psi \cos(k\vartheta),$$

$$\sum_{j=0}^{p-1} T_3 \cdot l_j^0 [(\Delta\varphi_j)_k - (\delta_{0j} + \delta_{1j})(\Delta\widetilde{\varphi}_j)_k] + \sum_{j=0}^{1} T_3 \cdot l_j^0 (\Delta\widetilde{\varphi}_j)_{k+1} = -\chi \cos\psi,$$

$$\sum_{j=2}^{p-1} T_1 \cdot A(j-1)(\Delta\varphi_j)_k + \sum_{j=0}^{1} c_j(\Delta\widetilde{\varphi}_j)_{k+1} = -\chi(d/2)\sin\psi \cos(k\vartheta),$$

$$\sum_{j=2}^{p-1} T_2 \cdot A(j-1)(\Delta\varphi_j)_k - \sum_{j=0}^{1} f_j(\Delta\widetilde{\varphi}_j)_{k+1} = \chi(d/2)\sin\psi \sin(k\vartheta) + \chi\rho\cos\psi,$$

$$\sum_{j=2}^{p-1} T_3 \cdot A(j-1)(\Delta\varphi_j)_k + \sum_{j=0}^{1} e_j(\Delta\widetilde{\varphi}_j)_{k+1} = -\chi\rho \sin\psi \cos(k\vartheta) \quad (k = 0, ..., n-1), \tag{15}$$

with

$$a_j = \cos\vartheta T_1 \cdot l_j^0 - \sin\vartheta T_2 \cdot l_j^0,$$
$$b_j = \sin\vartheta T_1 \cdot l_j^0 + \cos\vartheta T_2 \cdot l_j^0,$$
$$c_j = d\sin\vartheta T_1 \cdot l_j^0 + d\cos\vartheta T_2 \cdot l_j^0 - \rho\sin\vartheta T_3 \cdot l_j^0,$$
$$f_j = d\cos\vartheta T_1 \cdot l_j^0 - d\sin\vartheta T_2 \cdot l_j^0 + \rho(1 - \cos\vartheta) T_3 \cdot l_j^0,$$
$$e_j = \rho[\sin\vartheta T_1 \cdot l_j^0 - (1 - \cos\vartheta) T_2 \cdot l_j^0].$$

It should be pointed out that the set of equations (15) describes the variations of the rotation angles in the kth minor structural unit. Each minor structural unit (within the major one) may be characterized by its own set of values $\{\Delta\varphi_j\}_k$ as well as $\{\Delta\widetilde{\varphi}_j\}_k$. Thus, in order to evaluate the conformational changes of a helical macromolecule deformed into a coiled-coil, we have to solve the set of equations (15) for all values $k = 0, 1, ..., n-1$.

If $p = 1$ or 2, the set of eqautions (15) are incompatible. This means that a minor structural unit of the superhelix cannot consist of one or two bonds. Indeed, according to our model the angle of rotation ϑ was assumed to be constant in the distorted initial helix and a chain forms superhelix solely due to the periodical shortening and lengthening of the vector b_k. But for a chain with $p = 1$ or 2 the vector length $|b_k|$ is constant for any rotation angle (at $\{l_i\}$ = const, $\{\theta_i\}$ = const). If, nevertheless, a macromolecule with a structural unit of one or two bonds forms a superhelix then the minor structural units of the superhelix have to consist of several structural units of the original helix.

The equations (15) are invariant relative to the transformations

$$\chi, \psi \to -\chi, -(\pi-\psi). \tag{16}$$

This is equal to a reversal of the directions of the axes $(y, z)_k, (\eta, \zeta)_k, (y'', z'')_k$ and confirms the equivalence of both directions in the macromolecule.

5. Results and discussion

The equations (15) are used to obtain the variations of the rotation angles for the DNA double-helix forming a coiled-coil. For DNA $p = 6$ and the number of unknowns in the set (15) is greater than the number of equations. In this case the set of equations (15) is compatible but indefinite. The variables $(\Delta\widetilde{\varphi}_0)_{k+1}$ and $(\Delta\widetilde{\varphi}_1)_{k+1}$ were taken as arbitrary. Varying them, we found solutions of the set (15) and selected among them such solution which lead to the minimum values of other unknowns $(\Delta\varphi_j)_k$ $(j = 0, 1, ..., p-1)$. For simplicity $(\Delta\widetilde{\varphi}_0)_0$ and $(\Delta\widetilde{\varphi}_1)_0$ were assumed to be equal to zero. Conformational characteristics of the DNA in B-form were taken from Arnott's paper [9]. For the superhelix parameters we take the identity period $D = 120$ Å and the radius of helix $R = 50$ and 60 Å. To find the effect of the angle of inclination of the major helix $(\pi/2-\psi)$ on the form of the solution, we take various values of the number of turns in the identity period M and, consequently, N $[N^2(nd)^2 = M^2(2\pi R)^2 + D^2]$ at fixed D and R. The limiting case $M \to \infty$ $(\pi/2 - \psi \to 0)$ for which the axis of the ordinary helix is deformed into a circle and the superhelix degenerates into a tore, was also considered. The set of equations (15) was solved by the Gauss

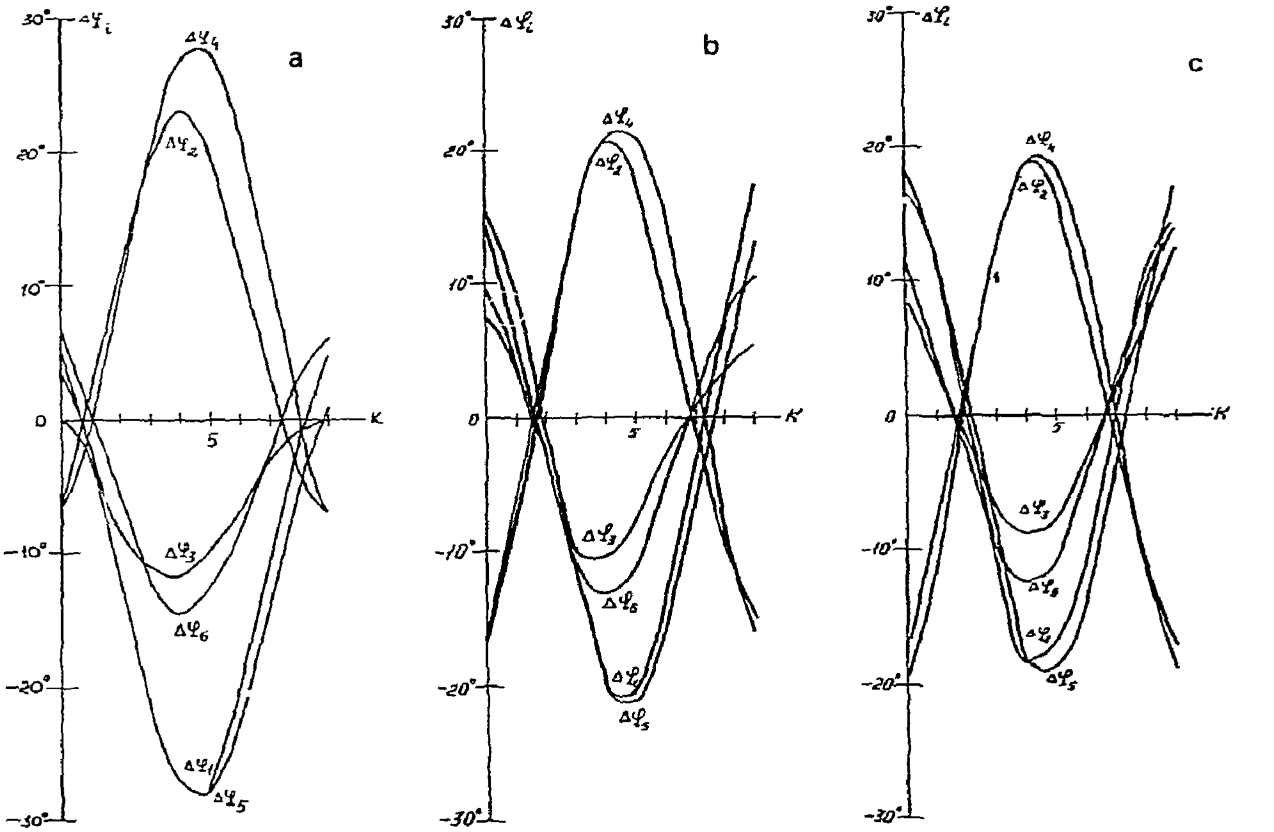

Fig. 3. Distortion of the rotation angles $\{\Delta\varphi_j\}_k$ $(j = 0, ..., 5)$ of the DNA in B-form in the superhelix formation. k is the number of the minor structural unit $(k = 0, ..., n-1)$. The values of parameters are as follows: $R = 50$ Å, $D = 120$ Å, $\rho = 9$ Å, $d = 3.4$ Å, $\vartheta = 35.5°$. (a) $M = 1$ $(\psi = 69.3°, \chi = 3.6°, nN = 100)$; (b) $M = 5$ $(\psi = 85.6°, \chi = 3.9°, nN = 460)$; (c) $M \to \infty$ $(\psi \to \pi/2, \chi = 3.9°, nN = 100)$.

method. Results obtained for the case where a right-handed helix of DNA is deformed into a left-handed major helix are shown in fig. 3 for $M = 1, 5$ and ∞.

As can be seen in fig. 3, within the major structural unit the changes of the rotation angles strongly depend on the number of the minor structural unit k. This means that different segments of the major structural unit are deformed differently and, therefore, each minor structural unit is characterized by its own set of rotation angles $\{\varphi_j\}_k$ $(j = 0, 1, ..., p-1)$. The middle segments of the major structural unit ($k = 4$ and 5) are subjected to the maximum deformation: they experience the largest compression (b_k is shortened) and $|\Delta\varphi_j|_k$ reach their maximum value.

A decrease in the angle of inclination of the major helix $(\pi/2 - \psi)$ (i.e., an increase in the number of turns M in the identity period approaching the limiting case of a tore) leads to a considerable deformation of the terminal segments ($k = 0$ or 9) of the major structural unit: they experience the largest extension (b_k is lengthened). An increase of $|\Delta\varphi_j|_k$ for $k = 0$ or 9 is accompanied by a decrease of $|\Delta\varphi_j|_k$ for $k = 4$ and 5 and in the limit their values are almost coincident.

The variations shown in fig. 3 are obtained for M = 1 and 5. The resulting curves for intermediate values of M are similar. Their extremes lie between the corresponding values $|(\Delta\varphi_j)_k|_{max}$ shown in fig. 3. At M = 2, 3 and, consequently, Nn = 190–280, the "mean deformation" of the major structural unit reaches a certain minimum (the deformation of the middle segments diminishes but that of the terminal segments does not increase yet sufficiently). Therefore, the superhelix with such "external" characteristics may be more favorable than any other one with fixed D and R.

With increasing radius of the major helix R the general picture remains the same but $(\Delta\varphi_j)_k$ slightly diminish.

Deviations of the rotation angles from their values in B-form of DNA are rather small and the maximum ones do not exceed 28° (at $R = 50$ and $M = 1$). It is interesting to compare the values obtained with the variations of the rotation angles in different forms of DNA [10] (see table 2). It can be seen that on the whole the values $\{\Delta\varphi_j\}_{max}$ obtained do not exceed those corresponding to the transition from the B-form to another one and have the same sign as the values for the B → C transition.

The results of this work demonstrate that the DNA backbone, in general, can form a superhelix owing to the slight distortion of the rotation angles. It is obvious that the calculation of the superhelix energy must be carried out for a detailed analysis.

It should also be pointed out that, because the maximum distortion of the rotation angles in the process of deformation reaches 28° and may be even greater (at $R < 50$ Å), the linear approximation in eqs. (2), (3), (A.4), (A.5), (A.6), (5), (7), (9), (11) may be not quite correct. Therefore, values $\{\Delta\varphi_j\}_k$ derived from the set of equations (15) should be treated as the zeroth solution of the exact equations (2) and (3); they may be used to find new solutions of higher approximation.

Another interesting aspect should be mentioned. There are experimental data showing that besides the chromatine structural units of 200 base pairs of DNA, there exists a shorter periodicity with a structural unit

Table 2
Deviations of the rotation angles in different helical forms of DNA from their values in B-form

The axis of internal rotation	Rotation angle in B-form	Deviations $\Delta\varphi_i$ of the rotation angles			
		B-form in superhelix	C-form	T-form	A-form
$\varphi°$: O_1–P	264.4	−27.9	−52.7	10.6	48.7
$\omega°$: C_3–O_1	154.7	20.5	56.7	−18.7	23.4
$\sigma°$: C_4–C_3	156.5	−9.5	−15.9	−10.5	−73.3
$\xi°$: C_5–C_4	36.4	27.7	11.3	49.6	8.9
$\theta°$: O_4–C_5	213.5	−28.0	−70.1	9.5	−5.1
$\psi°$: P–O_4	313.9	−12.9	0.7	−26.9	−38.6

of about 10 (or 20) base pairs [11]. The existence of the shorter periodicity may be easily understood from the superhelix model if we assume that the "major" periodicity is connected with the identity period of the major helix and the "minor" periodicity corresponds to the identity period of the minor helix. According to the foregoing, these data can be attributed to the superhelix with R = 50 Å, D = 120 Å and M = 2. Indeed, in this case the "major" periodicity would be connected with N = 19 major structural units, containing therefore, Nn = 190 base pairs and the "minor" periodicity would be connected with one major structural unit, containing 10 minor units, i.e. 10 base pairs.

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Appendix I

In eqs. (2) and (3) the transformation matrix $\hat{\mathbf{N}}$ may be written as follows

$$\hat{\mathbf{N}}_k = (\hat{\mathbf{Q}}_k \hat{\mathbf{P}}_k)^{-1} \hat{\mathbf{Q}}_{k+1} \hat{\mathbf{P}}_{k+1}, \tag{A.1}$$

where $\hat{\mathbf{P}}_j$ is the matrix of transformation from the $(\xi, \eta, \zeta)_j$ to the $(x, y, z)_j$ coordinate system:

$$\hat{\mathbf{P}}_j = \begin{pmatrix} \cos(j\vartheta) & -\sin(j\vartheta) & 0 \\ \sin(j\vartheta) & \cos(j\vartheta) & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{A.2}$$

$\hat{\mathbf{Q}}_j$ is the matrix of transformation from coordinate system $(x, y, z)_j$ to the fixed coordinate system (X, Y, Z)

$$\hat{\mathbf{Q}}_j = \begin{pmatrix} \cos(j\chi) & \cos\psi \sin(j\chi) & -\sin\psi \sin(j\chi) \\ -\sin(j\chi) & \cos\psi \cos(j\chi) & -\sin\psi \cos(j\chi) \\ 0 & \sin\psi & \cos\psi \end{pmatrix}. \tag{A.3}$$

Introducing eqs. (A.2) and (A.3) in eq. (A.1) we have

$$\hat{\mathbf{N}}_k = \begin{pmatrix} \cos\vartheta - (1-\cos\chi)a_{11} + c_1 \sin\chi & -\sin\vartheta + (1-\cos\chi)a_{12} + c_2 \sin\chi & a_{13} \sin\psi \\ \sin\vartheta + (1-\cos\chi)a_{21} - c_2 \sin\chi & \cos\vartheta - (1-\cos\chi)a_{22} + c_1 \sin\chi & a_{23} \sin\psi \\ a_{31} \sin\psi & a_{32} \sin\psi & \cos\chi + \cos^2\psi(1-\cos\chi) \end{pmatrix}, \tag{A.4}$$

where

$$a_{11} = \cos[(k+1)\vartheta] \cos(k\vartheta) + \cos^2\psi \sin[(k+1)\vartheta] \sin(k\vartheta),$$

$$a_{12} = \sin[(k+1)\vartheta] \cos(k\vartheta) - \cos^2\psi \cos[(k+1)\vartheta] \sin(k\vartheta),$$

$$a_{13} = -\sin\chi \cos(k\vartheta) + \cos\psi(1-\cos\chi) \sin(k\vartheta),$$

$$a_{21} = \cos[(k+1)\vartheta] \sin(k\vartheta) - \cos^2\psi \sin[(k+1)\vartheta] \cos(k\vartheta),$$

$$a_{22} = \sin[(k+1)\vartheta] \sin(k\vartheta) + \cos^2\psi \cos[(k+1)\vartheta] \cos(k\vartheta),$$

$$a_{23} = \sin\chi \sin(k\vartheta) + \cos\psi(1-\cos\chi) \cos(k\vartheta),$$

$$a_{31} = \sin\chi \cos[(k+1)\vartheta] + \cos\psi(1-\cos\chi) \sin[(k+1)\vartheta],$$

$$a_{32} = -\sin\chi \sin[(k+1)\vartheta] + \cos\psi(1-\cos\chi) \cos[(k+1)\vartheta],$$

$c_1 = \cos\psi \sin\vartheta, \quad c_2 = \cos\psi \cos\vartheta.$

The transformation matrix $\hat{F}_k$ can be expressed as

$$\hat{F}_k = (\hat{Q}_k \hat{P}_k)^{-1} = \begin{pmatrix} f_{11} & f_{12} & \sin\psi \sin(k\vartheta) \\ f_{21} & f_{22} & \sin\psi \cos(k\vartheta) \\ -\sin\psi \sin(k\chi) & -\sin\psi \cos(k\chi) & \cos\psi \end{pmatrix}, \tag{A.5}$$

where

$$f_{11} = \cos(k\chi)\cos(k\vartheta) + \cos\psi \sin(k\chi)\sin(k\vartheta), \qquad f_{12} = -\sin(k\chi)\cos(k\vartheta) + \cos\psi \cos(k\chi)\sin(k\vartheta),$$

$$f_{21} = -\cos(k\chi)\sin(k\vartheta) + \cos\psi \sin(k\chi)\cos(k\vartheta), \quad f_{22} = \sin(k\chi)\sin(k\vartheta) + \cos\psi\cos(k\chi)\cos(k\vartheta).$$

The components of the vector $(Oo_{k+1} - Oo_k)$ in the fixed coordinate system (X, Y, Z) are

$$Oo_{k+1} - Oo_k = \begin{pmatrix} R\{\cos[(k+1)\chi] - \cos(k\chi)\} \\ -R\{\sin[(k+1)\chi] - \sin(k\chi)\} \\ H \end{pmatrix}. \tag{A.6}$$

Appendix 2

(1) In eqs. (A.4), (A.5) and (A.6) expand the elements of matrices $\hat{N}_k$, $\hat{F}_k$ and vector $(Oo_{k+1} - Oo_k)$ into a power series of the parameter χ and retain only linear terms. Then

$$\hat{N}_k = \hat{N} + \chi \hat{n}_k, \tag{A.7}$$

where the matrix $\hat{N}$ is just the same as the corresponding matrix in ref. [8]

$$\hat{N} = \begin{pmatrix} \cos\vartheta & -\sin\vartheta & 0 \\ \sin\vartheta & \cos\vartheta & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{A.8}$$

and

$$\hat{n}_k = \begin{pmatrix} \cos\psi \sin\vartheta & \cos\psi\cos\vartheta & -\sin\psi\cos(k\vartheta) \\ -\cos\psi\cos\vartheta & \cos\psi \sin\vartheta & \sin\psi \sin(k\vartheta) \\ \sin\psi \cos[(k+1)\vartheta] & -\sin\psi\sin[(k+1)\vartheta] & 0 \end{pmatrix}. \tag{A.9}$$

$$\hat{F}(Oo_{k+1} - Oo_k) - (\hat{E} - \hat{N}_k)\rho = \hat{T}^{-1}b + \chi m_k. \tag{A.10}$$

$\hat{T}^{-1}b$ coincides with the analogous expression in ref. [8]:

$$\hat{T}^{-1}b = \begin{pmatrix} -\rho(1 - \cos\vartheta) \\ \rho \sin\vartheta \\ d \end{pmatrix} \tag{A.11}$$

and

$$m_k = \begin{pmatrix} -(d/2)\sin\psi\cos(k\vartheta) + \rho\cos\psi\sin\vartheta \\ (d/2)\sin\psi\sin(k\vartheta) - \rho\cos\psi\cos\vartheta \\ \rho\sin\psi\cos[(k+1)\vartheta] \end{pmatrix}. \tag{A.12}$$

(2) Taking into consideration eq. (14), expand the components of the matrix (7) into a power series of $(\Delta\varphi_j)_k$ and retain only linear terms. Then

$$(\hat{S}_j)_k = \hat{S}_j + (\Delta\varphi_j)_k \hat{L}_j, \tag{A.13}$$

where

$$\hat{L}_j = \begin{pmatrix} \cos\alpha_j\sin\varphi_j & \cos\varphi_j & \sin\alpha_j\sin\varphi_j \\ -\cos\alpha_j\cos\varphi_j & \sin\varphi_j & -\sin\alpha_j\cos\varphi_j \\ 0 & 0 & 0 \end{pmatrix}. \tag{A.14}$$

Substitution of eq. (A.13) in eq. (5) gives

$$\hat{S}_k = \prod_{j=0}^{p-1} \hat{S}_j + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \prod_{t=0}^{j-1} \hat{S}_t \hat{L}_j \prod_{t=j+1}^{p-1} \hat{S}_t, \tag{A.15}$$

which can be rewritten as follows

$$\hat{S}_k = \hat{S} + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \prod_{t=0}^{j-1} \hat{S}_t \hat{L}_j \hat{S}_j^{-1} \left(\prod_{t=0}^{j-1} \hat{S}_t \right)^{-1} \hat{S} = \hat{S} + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \hat{S}(j-1)\hat{K}\hat{S}^{-1}(j-1)\hat{S}, \tag{A.16}$$

where

$$\hat{S}(j-1) \equiv \prod_{t=0}^{j-1} \hat{S}_t, \tag{A.17}$$

$$\hat{K} \equiv \hat{L}_j \hat{S}_j^{-1} = \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{A.18}$$

and finally we have

$$\hat{S}_k = \hat{S} + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \hat{W}_{j-1} \hat{S}, \tag{A.19}$$

where

$$\hat{W}_{j-1} \equiv \hat{S}(j-1)\hat{K}\hat{S}^{-1}(j-1) = \begin{pmatrix} 0 & -[\hat{S}(j-1)]_{33} & [\hat{S}(j-1)]_{23} \\ [\hat{S}(j-1)]_{33} & 0 & -[\hat{S}(j-1)]_{13} \\ -[\hat{S}(j-1)]_{23} & [\hat{S}(j-1)]_{13} & 0 \end{pmatrix}. \tag{A.20}$$

At $j = 0$, $\hat{W}_{-1} = \hat{K}$. In the same way

$$\hat{T}_k = \hat{T} + \sum_{j=0}^{1} (\Delta\tilde{\varphi}_j)_k \hat{W}_{j-1} \hat{T}, \tag{A.21}$$

$$\hat{T}_k^{-1} = \hat{T}^{-1} - \sum_{j=0}^{1} (\Delta\tilde{\varphi}_j)_k \hat{T}^{-1} \hat{W}_{j-1}. \tag{A.22}$$

For b_k we have the following expression

$$b_k = b + \sum_{t=0}^{p-1} \sum_{j=0}^{t} (\Delta\varphi_j)_k \hat{W}_{j-1} \prod_{f=0}^{t} \hat{S}_f l_{t+1} = b + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \hat{W}_{j-1} \sum_{t=j}^{p-1} \left(\prod_{f=0}^{t} S_f\right) l_{t+1}, \quad (A.23)$$

and taking into account that

$$\hat{W}_{j-1} \prod_{f=0}^{j-1} S_f l_j = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix},$$

we have

$$b_k = b + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \hat{W}_{j-1} [b - b(j-1)], \quad (A.24)$$

where

$$b(j-1) = \sum_{t=0}^{j-2} \left(\prod_{f=0}^{t} S_f\right) l_{t+1}. \quad (A.25)$$

Substituting eqs. (A.19), (A.21) and (A.22) in the right side of eq. (2) and using the condition $\hat{N} = \hat{T}^{-1}\hat{S}\hat{T}$ we find

$$\hat{T}_k^{-1}\hat{S}_k\hat{T}_{k+1} = \hat{T}^{-1}\hat{S}\hat{T} + \sum_{j=0}^{p-1} [(\Delta\varphi_j)_k - (\delta_{0j}+\delta_{1j})(\Delta\tilde{\varphi}_j)_k]\, \hat{T}^{-1}\hat{W}_{j-1}\hat{T}\hat{N} + \sum_{j=0}^{1} (\Delta\tilde{\varphi}_j)_{k+1} \hat{N}\hat{T}^{-1}\hat{W}_{j-1}\hat{T}. \quad (A.26)$$

It can be readily shown that

$$\hat{T}^{-1}\hat{W}_{j-1}\hat{T} = \begin{pmatrix} 0 & -T_3 \cdot l_j^0 & T_2 \cdot l_j^0 \\ T_3 \cdot l_j^0 & 0 & -T_1 \cdot l_j^0 \\ -T_2 \cdot l_j^0 & T_1 \cdot l_j^0 & 0 \end{pmatrix}, \quad (A.27)$$

where

$$l_j^0 \equiv \hat{S}(j-1)(l_j/|l_j|), \quad l_0^0 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \quad (A.28)$$

$$\hat{T} \equiv (T_1\ T_2\ T_3).$$

Using eq. (A.8) we finally obtain

$$\hat{T}_k^{-1}\hat{S}_k\hat{T}_{k+1} = \hat{N} + \sum_{j=0}^{p-1} [(\Delta\varphi_j)_k - (\delta_{0j}+\delta_{1j})(\Delta\tilde{\varphi}_j)_k]\, \hat{B}_j + \sum_{j=0}^{1} (\Delta\tilde{\varphi}_j)_{k+1} \hat{C}_j, \quad (A.29)$$

where

$$\hat{B}_j \equiv (\hat{T}^{-1}\hat{W}_{j-1}\hat{T})\hat{N} = \begin{pmatrix} -\sin\vartheta\, T_3 \cdot l_j^0 & -\cos\vartheta\, T_3 \cdot l_j^0 & T_2 \cdot l_j^0 \\ \cos\vartheta\, T_3 \cdot l_j^0 & -\sin\vartheta\, T_3 \cdot l_j^0 & -T_1 \cdot l_j^0 \\ -\cos\vartheta\, T_2 \cdot l_j^0 + \sin\vartheta\, T_1 \cdot l_j^0 & \sin\vartheta\, T_2 \cdot l_j^0 + \cos\vartheta\, T_1 \cdot l_j^0 & 0 \end{pmatrix}, \quad (A.30)$$

$$\hat{\mathrm{C}}_j \equiv \hat{\mathrm{N}}(\hat{\mathrm{T}}^{-1}\hat{\mathrm{W}}_{j-1}\hat{\mathrm{T}}) = \begin{pmatrix} -\sin\vartheta\, T_3\cdot l_j^0 & -\cos\vartheta\, T_3\cdot l_j^0 & \sin\vartheta\, T_1\cdot l_j^0 + \cos\vartheta\, T_2\cdot l_j^0 \\ \cos\vartheta\, T_3\cdot l_j^0 & -\sin\vartheta\, T_3\cdot l_j^0 & \sin\vartheta\, T_2\cdot l_j^0 - \cos\vartheta\, T_1\cdot l_j^0 \\ -T_2\cdot l_j^0 & T_1\cdot l_j^0 & 0 \end{pmatrix}. \tag{A.31}$$

Substitute eqs. (A.22) and (A.24) in the right-handside of eq. (3). Then

$$\hat{\mathrm{T}}_k^{-1} b_k = \hat{\mathrm{T}}^{-1} b - \sum_{j=0}^{1} (\Delta\widetilde{\varphi}_j)_k \hat{\mathrm{T}}^{-1}\hat{\mathrm{W}}_{j-1} b + \sum_{j=0}^{p-1} (\Delta\varphi_j)_k \hat{\mathrm{T}}^{-1}\hat{\mathrm{W}}_{j-1}[b - b(j-1)]. \tag{A.32}$$

It can be shown that

$$\hat{\mathrm{T}}^{-1}\hat{\mathrm{W}}_{j-1} b(j-1) = \begin{pmatrix} T_1\cdot A(j-1) \\ T_2\cdot A(j-1) \\ T_3\cdot A(j-1) \end{pmatrix}, \tag{A.33}$$

where

$$A(j-1) = [l_j^0 \times b(j-1)]. \tag{A.34}$$

Using eq. (A.11) we have

$$\hat{\mathrm{T}}_k^{-1} b_k = \hat{\mathrm{T}}^{-1} b + \sum_{j=0}^{p-1} [(\Delta\varphi_j)_k - (\delta_{0j}+\delta_{1j})(\Delta\widetilde{\varphi}_j)_k] D_j, \tag{A.35}$$

where

$$D_j = \begin{pmatrix} -\rho\sin\vartheta\, T_3\cdot l_j^0 + d T_2\cdot l_j^0 - (1-\delta_{0j})(1-\delta_{1j}) T_1\cdot A(j-1) \\ -\rho(1-\cos\vartheta) T_3\cdot l_j^0 - d T_1\cdot l_j^0 - (1-\delta_{0j})(1-\delta_{1j}) T_2\cdot A(j-1) \\ \rho(1-\cos\vartheta) T_2\cdot l_j^0 + \rho\sin\vartheta\, T_1\cdot l_j^0 - (1-\delta_{0j})(1-\delta_{1j}) T_3\cdot A(j-1) \end{pmatrix}. \tag{A.36}$$

Substituting eqs. (A.7), (A.10), (A.29) and (A.35) in eqs. (2) and (3) and equaling the respective elements on both sides of eqs. (2) and (3), we obtain the set of linear algebraic equations (15).

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