

- BRUCE, A. D. & SCHNEIDER, T. (1977). *Phys. Rev. B*, **16**, 3991–3997.
- BULOUP, A., RIDOU, C., ROUSSEAU, M., NOUET, J. & HEWAT, A. (1980). *J. Phys.* **41**, 87–96.
- COCHRAN, W. (1960). *Adv. Phys.* **9**, 387–423.
- COWLEY, R. A. (1980). *Adv. Phys.* **29**, 1–110.
- DAWSON, B., HURLEY, A. C. & MASLEN, V. W. (1967). *Proc. R. Soc. London Ser. A*, **298**, 289–306.
- FUJII, Y., HOSHINO, S., YAMADA, Y. & SHIRANE, G. (1974). *Phys. Rev. B*, **9**, 4549–4559.
- HUTTON, J., NELMES, R. J., MEYER, G. M. & EIRIKSSON, V. R. (1979). *J. Phys. C*, **12**, 5393–5410.
- JOHNSON, C. E. (1970). *Thermal Neutron Diffraction*, edited by B. T. M. WILLIS. Oxford Univ. Press.
- LANDAU, L. D. & LIFSHITZ, E. M. (1958). *Statistical Physics*. London: Pergamon.
- MAIR, S. L. (1982). *J. Phys. C*, **15**, 25–36.
- MAIR, S. L., BARNEA, Z., COOPER, M. J. & ROUSE, K. D. (1974). *Acta Cryst.* **A30**, 806–813.
- MEGAW, H. D. (1969). *Acta Cryst.* **B25**, 1516–1517.
- MEISSNER, G. & BINDER, K. (1975). *Phys. Rev. B*, **12**, 3948–3955.
- POUGET, J. P., SHAPIRO, S. M. & NASSAU, K. (1979). *J. Phys. Chem. Solids*, **40**, 267–277.
- ROBERTO, J. B., BATTERMAN, B. W. & KEATING, D. T. (1974). *Phys. Rev. B*, **9**, 2590–2599.
- SAKATA, M., HARADA, J., COOPER, M. J. & ROUSE, K. D. (1980). *Acta Cryst.* **A36**, 7–15.
- VOGT, K., PRANDL, W. & REICHARDT, W. (1981). *Z. Phys.* **B41**, 321–328.
- WILLIS, B. T. M. & PRYOR, A. W. (1975). *Thermal Vibrations in Crystallography*. Cambridge Univ. Press.

Acta Cryst. (1982). **A38**, 796–799

The Fourier Transform of Actin and Other Helical Systems With Cumulative Random Angular Disorder

BY E. H. EGELMAN

Graduate Program in Biophysics, Brandeis University, Waltham, MA 02254, USA

AND D. J. DEROSIER

Biology Department and Rosenstiel Basic Medical Sciences Research Center, Brandeis University, Waltham, MA 02254, USA

(Received 15 February 1982; accepted 11 May 1982)

Abstract

A general class of helical disorder exists which can be described by cumulative random angular motions of subunits. This disorder affects layer-line intensities and widths by a factor proportional to n^2 , the square of the order of the layer line. The result explains several features of actin and polytetrafluoroethylene (Teflon) transforms, and may be relevant to other helical systems.

We have described the angular motions of subunits in the F-actin helix (Egelman, Francis & DeRosier, 1982) based upon image analysis of electron micrographs. In this paper we will present an analytic treatment of the effect of this form of disorder on the transform of such a structure. Because this treatment appears to explain features of other disordered systems (such as Teflon above the 292 K transition), we believe that it may be applicable to many helical structures.

Whereas the subunit positions in an ideal helix can be described by

$$r_j = r_0$$

$$z_j = j \Delta z$$

$$\psi_j = \psi_{j-1} + \Delta\psi = j \Delta\psi,$$

we will deal with a particular form of disordered helix where subunit positions are described by

$$r_j = r_0$$

$$z_j = j \Delta z$$

$$\psi_j = \psi_{j-1} + \Delta\psi + \delta_j = j \Delta\psi + \sum_{k=1}^j \delta_k. \quad (1)$$

Fig. 1 shows a model of a helix described by these equations, and Table 1 contains the first ten values of δ_j for one of the filaments in Fig. 1.

The recursive relation in (1) is nothing more than a correlated random walk in ψ , and can be parameterized in terms of the first moment and the square root of the second moment of the distribution of δ_j 's:

$$\langle \delta_j \rangle; \langle \delta_j^2 \rangle^{1/2} = \delta_{1,m,n}, \dots$$

We will define $\langle \delta_j \rangle \equiv 0$.

Thus, for a given ψ_j , we will expect to find $\psi_{j+1} = \psi_j + \Delta\psi \pm \delta_{r.m.s.}$; $\psi_{j+2} = \psi_j + 2\Delta\psi \pm 2^{1/2}\delta_{r.m.s.}$; etc. When $\delta_{r.m.s.}$ is small with respect to $\Delta\psi$, angular

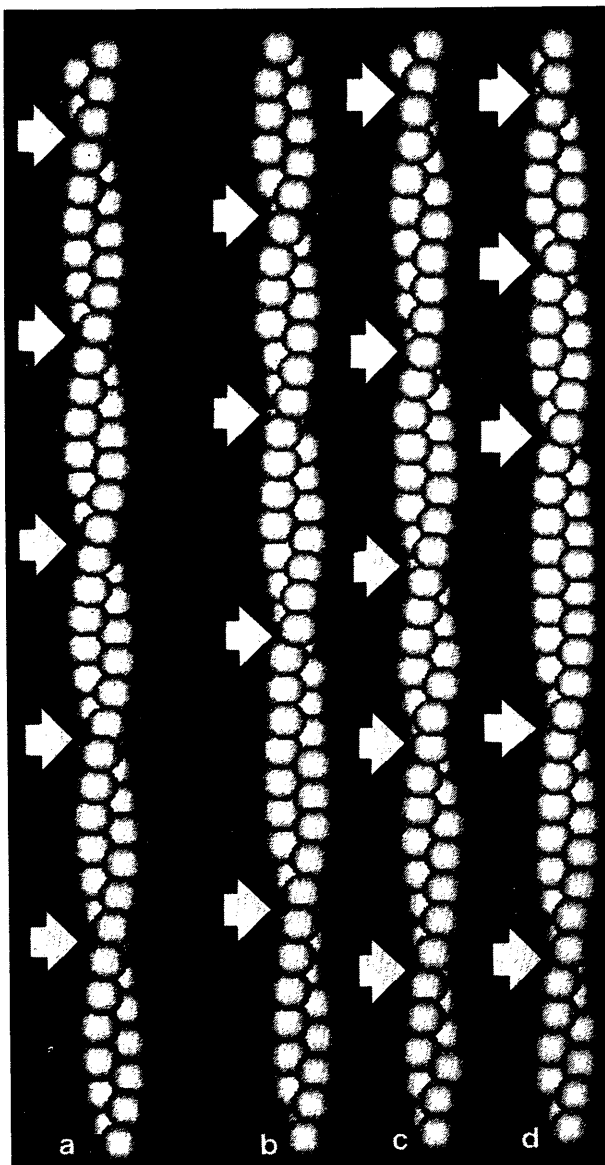


Fig. 1. A computer-generated model shows how cumulative angular disorder affects the appearance of a helical fiber. Spheres have been arranged according to the helical geometry of actin (approximately 13 subunits in six turns of a left-handed 59 Å pitch helix). In (a), the placement of subunits is regular. In (b)–(d), the angular rotation between subunits has an r.m.s. fluctuation of 10° , and these fluctuations have a Gaussian distribution. In all filaments shown, the axial rise per subunit is fixed, and all four filaments begin at the bottom with their first subunit in the same angular position. Because the deviations of the subunit angular positions are cumulative, the points where the right-handed two-start helices cross over (marked by the arrows) have been almost randomized in (b)–(d).

correlation will be maintained over many subunits. As $\delta_{r.m.s.}$ grows with respect to $\Delta\psi$, the correlation length will obviously fall.

The transform of both a perfect helix and our disordered helix can be understood in terms of a product of the transform of an individual subunit multiplied by the transform of the helical lattice. In deriving the Fourier transform of the disordered helix, we can ignore the radial component, since this disorder will not affect it. The transform of the remaining axial and azimuthal components of the helix generates a sampling function $C(n, Z)$ which gives rise to layer lines. Any given layer line will have an order n associated with it which is related to the azimuthal symmetry of the corresponding helix ($n = 1$ for a one-start, $n = 2$ for a two-start, etc.). We will calculate this sampling function and follow the standard convention where $Z = l/c$; l = layer-line number, c = helical repeat. For the ideal helix, we have

$$C(n, Z) = \sum_{j=1}^N \exp[i(-n\psi_j + 2\pi Z z_j)] \quad (2)$$

using

$$\begin{aligned} \psi_j &= j \Delta\psi \\ z_j &= j \Delta z = j p \Delta\psi / 2\pi, \end{aligned}$$

where p is the pitch of a one-start helix,

$$\begin{aligned} C(n, Z) &= \sum_{j=1}^N \exp[i(-nj\Delta\psi + jZp\Delta\psi)] \\ &= \sum_{j=1}^N \exp[i(j\Delta\psi(pZ - n))]. \end{aligned}$$

The corresponding $C(n, Z)$ for the disordered helix described by (1) is

$$\begin{aligned} C(n, Z) &= \sum_{j=1}^N \exp \left[i \left(-nj\Delta\psi - n \sum_{k=1}^j \delta_k + 2\pi Z z_j \right) \right] \\ C(n, Z) &= \sum_{j=1}^N \exp \left[i \left(j\Delta\psi(pZ - n) - n \sum_{k=1}^j \delta_k \right) \right]. \quad (3) \end{aligned}$$

The exact solution of this equation depends upon the detailed knowledge of the δ_k 's, including both their individual values as well as their sequence. However, using statistical methods, we can solve for the expectation value, $\langle C(n, Z) \rangle$ in terms of another expectation value, $\langle \delta_k^2 \rangle^{1/2}$. From (2) we can see that for a given n there will exist a family of $Z_{0,n}$'s (layer lines) such that

$$\Delta\psi(pZ_{0,n} - n) = 2\pi m; \quad m = 0, \pm 1, \pm 2, \dots$$

It can easily be shown that the disorder term in (3) will not shift the expectation value of these $Z_{0,n}$'s. This follows from the expectation value of the sum over the δ_k 's being equal to zero. Therefore, the expectation value for the layer-line peak will always be at the same position it would be in the absence of disorder.

Thus, at the expected layer-line peaks, (3) reduces to

$$C(n, Z_{0,n}) = \sum_{i=1}^n \exp \left[-in \sum_{k=1}^i \delta_k \right]. \quad (4)$$

when $\delta_{r.m.s.} = 0$, $|C(n, Z)| = N$.

For the more general situation ($\delta_{r.m.s.} \neq 0$) it is helpful to rewrite the double summation in (4) as a single sum:

$$C(n, Z_{0,n}) = \sum_{j=1}^n \exp(-i\beta_j), \quad (5)$$

where

$$\langle (\beta_j - \beta_{j+1})^2 \rangle^{1/2} = n\delta_{r.m.s.}$$

The magnitude of this summation will build as a correlated random walk. That is, for $n\delta_{r.m.s.}$ significantly less than 2π (for actin, we have experimentally determined $\delta_{r.m.s.}$ to be of the order of 10°), successive terms in the summation will involve the addition of vectors (whose angle is given by β) which are quite well correlated in angle. Thus, these terms will build as N , the number of steps. However, over many steps, the vectors will appear to be doing a random walk in the complex plane. In this limit, the summation should grow as $N^{1/2}$.

In Fig. 2, the summation of (5) is performed for the first ten subunits of Fig. 1, filament b . One can see that for $n = 3$ the correlation between steps in the complex plane begins to die off much more rapidly than for $n = 1$. While both curves ($n = 1$ and $n = 3$) have a random component in their growth, it can be seen that for $N = 10$ there is still a strong correlation in both between the first step and the tenth. This description of a correlated random walk is isomorphic with the expected end-to-end distance of a long filamentous polymer, which is bending in solution. Over a very long length the filament behaves as a random coil, while over short stretches the filament is best approximated as a relatively rigid rod. The mean squared end-to-end distance of such a flexible polymer can be written as (Landau & Lifschitz, 1958)

Table 1. Model values for cumulative disorder

j	δ_j ($^\circ$)	β_j ($n = 1$) ($^\circ$)	β_j ($n = 3$) ($^\circ$)
1	12.5	12.5	37.4
2	15.8	28.3	84.8
3	-15.4	12.9	38.6
4	-4.9	7.9	23.8
5	-7.8	0.1	0.4
6	6.3	6.4	19.2
7	-9.9	-3.5	-10.6
8	7.6	4.0	12.1
9	6.6	10.6	31.8
10	2.6	13.2	39.5

These are the values of δ_j for the first ten subunits of Fig. 1, filament b . The corresponding β_j 's are as defined in equation (5).

$$\langle R^2 \rangle = 2b^2 \left(\frac{L}{b} - 1 + e^{-L/b} \right),$$

where b is a correlation length and L is the contour length along the polymer. The expectation value of $|C(n, Z_{0,n})|^2$ will behave in the same manner as $\langle R^2 \rangle$, and we can therefore write

$$\langle |C(n, Z_{0,n})|^2 \rangle = \frac{a^2}{2} \left(\frac{2N}{a} - 1 + e^{-2N/a} \right), \quad (6)$$

where N is the number of subunits (analogous to L). We have redefined the bending correlation length b in terms of a diffraction correlation length a , $2b = a$, for later simplicity. We can define this new correlation length by once again referring to polymer bending, in which we find the relationship (Landau & Lifshitz, 1958)

$$e^{-L/b} = \langle \cos \theta(L) \rangle,$$

where θ is the angle between tangents to a filament at two points separated by the contour length L . We can now write a corresponding equation for our walk in the complex plane:

$$\langle \cos(n\delta_k) \rangle = e^{-2/a},$$

where the mean angular deviation after one subunit ($N = 1$) is $n\delta_{r.m.s.}$. For $n\delta_{r.m.s.} \ll 1$, we can expand this and we have

$$a = 4/n^2 \delta_{r.m.s.}^2.$$

The most unusual feature of this equation is that the correlation length varies for different layer lines. For example, we determined that for actin $\delta_{r.m.s.} = 10^\circ$. On the sixth layer line of actin where $n = -1$, $a_{-1} = 131$ subunits while on the first layer line, where $n = 2$, $a_2 = 33$ subunits.

It is important to note that the intensities of these layer lines will be differently affected. We can simplify

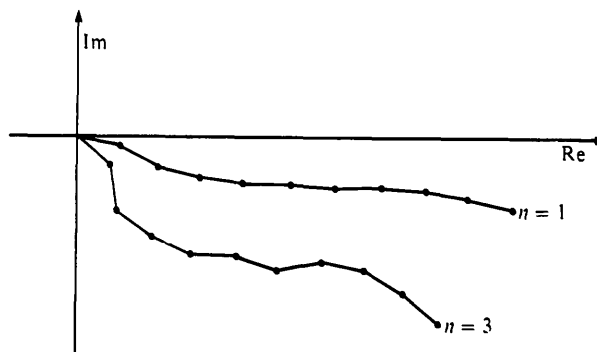


Fig. 2. The growth of the sampling function, $C(n, Z_{0,n})$, proceeds as a correlated random walk in the complex plane. We have plotted here the summation in equation (5) for the first ten subunits of Fig. 1, filament b . The values of δ_j and β_j for these subunits are given in Table 1.

(6) for the case where $N \gg a_n$, that is, the filaments we are diffracting from are longer than the correlation length:

$$\langle |C(n, Z_{0,n})|^2 \rangle = aN = 4N/n^2 \delta_{r.m.s.}^2.$$

Thus, when one is dealing with lengths significantly greater than the correlation length an important feature emerges from this equation. The peak layer-line intensities (except for meridional reflections which will be unaffected by this disorder) will be weighted by a factor of $1/n^2$. Further, since the *total* intensity of scattering will still be equal to N for both the ordered and disordered case, the width of a layer line in the meridional direction will be independent of N when $N \gg a$. Since the peak intensity is equal to $4N/n^2 \delta_{r.m.s.}^2$, the width will be equal to $n^2 \delta^2/4$ ($= 1/a$) in this regime. Thus the helical object diffracts as a mosaic of helical stretches of length a_n where the length is different for different layer lines.

All of these statistical predictions have been confirmed using Monte Carlo simulations of helical filaments whose geometry is described by (1). These simulations have involved up to one million subunits, and their results in terms of layer-line intensities and widths have been in excellent agreement with our analytical treatment.

The practical consequences of these features are quite significant for X-ray diffraction. As n increases, not only will the peak intensity be decreased, but the increased width of the layer line will help to make it indistinguishable from the background incoherent scatter.

We have dealt elsewhere (Egelman *et al.*, 1982) with how the predictions of this formalism successfully describe several features of the actin transform. We would like to point out here that this formalism is equally relevant for describing the unsampled fiber pattern of polytetrafluoroethylene after the 292 and 303 K transitions. It has been understood for a considerable time from X-ray diffraction (Clark & Muus, 1962*b*) and NMR (Hyndman & Origlio, 1960) that individual chains in polytetrafluoroethylene crystals undergo torsional motions after the 293 K transition and that these motions appear to be of even greater magnitude after the 303 K transition. A previous treatment (Clark & Muus, 1962*a*) of the effect of this disorder dealt only with the crystal Bragg reflections (that is, the sampled intensities on layer lines). We believe that our treatment extends this to explain the appearance of the unsampled continuous layer-line intensities arising from the disorder. Because the meridional reflections remain sharp after both of these transitions, it is reasonable to believe that our equation (1) provides a good description of the motions in the Teflon chains. The $n = 2$ ($l = 1$) continuous peak

intensity seems to disappear after the 292 K transition, while the $n = 1$ reflections on the sixth and seventh layer lines themselves weaken or disappear after the 303 K transition. This is exactly the prediction of our formalism, where the correlation length for all $n \neq 0$ falls as the temperature increases. For any given temperature, the correlation length for a layer line will be proportional to $1/n^2$.

That our equation (1) describes the disorder in Teflon fairly well is further supported by model-potential-energy calculations (DeSantis, Giglio, Liquori & Ripamonti, 1963; McMahon & McCullough, 1965; Bates, 1967; Bates & Stockmayer, 1968) for free filaments which yield a relatively shallow potential-energy well for rotations about the C—C bond. For $T = 300$ K, a shift of kT from the most stable bond angle (about 165°) appears to be equal to about $\pm 8^\circ$ (McMahon & McCullough, 1965, Fig. 2). These shifts can be mapped into the helical coordinates ψ and z . The absolute shift in $\Delta\psi$ (the angular rotation between subunits) is about the same as the shift in the bond angle, but the fractional change in $\Delta\psi$ for this amount of rotation is 15 times greater than the corresponding fractional change in the axial rise Δz of each subunit. Therefore, just like actin, almost all of the disorder is azimuthal.

Because equation (1) appears to provide a reasonable approximation of these two helical systems (actin and Teflon), it suggests that this form of disorder may be a general feature of other filamentous polymers where the number of connections between subunits is limited. The essential requirement for this formalism to be applicable is that the angular correlation for subunits must die off faster than the axial correlation. The helical structures of both actin and polytetrafluoroethylene appear to obey this requirement.

References

- BATES, T. W. (1967). *Trans. Faraday Soc.* **63**, 1825.
 BATES, T. W. & STOCKMAYER, W. H. (1968). *Macromolecules*, **1**, 12–24.
 CLARK, E. S. & MUUS, L. T. (1962*a*). *Z. Kristallogr.* **117**, 108–118.
 CLARK, E. S. & MUUS, L. T. (1962*b*). *Z. Kristallogr.* **117**, 119–127.
 DESANTIS, P., GIGLIO, E., LIQUORI, A. M. & RIPAMONTI, A. (1963). *J. Polym. Sci. Part A*, **1**, 1383–1404.
 EGELMAN, E. H., FRANCIS, N. & DEROSIER, D. J. (1982). *Nature (London)*, **298**, 131–135.
 HYNDMAN, D. & ORIGLIO, G. F. (1960). *J. Appl. Phys.* **31**, 1849–1852.
 LANDAU, L. & LIFSHITZ, E. M. (1958). *Statistical Physics*, pp. 479–482. London: Pergamon.
 MCMAHON, P. E. & MCCULLOUGH, R. L. (1965). *Trans. Faraday Soc.* **61**, 201–206.