

The Fourier Coefficients of Paracrystalline X-ray Diffraction*

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Theoretical expressions are presented for the Fourier coefficients, derived in terms of a linear 'ideal paracrystalline' lattice defined by Hosemann. These coefficients are related to the 'coordination statistic' of paracrystallites and their size distribution. The expressions have been used to review earlier Fourier transform analyses on 'Marlex 50' polyethylene and various heat-treated samples of petroleum coke and it has been established that these substances have a homogeneous paracrystalline disorder. Up-to-date results of the X-ray diffraction line-profile analyses of various paracrystalline substances are summarized and discussed.

Introduction

The theory of paracrystals (Hosemann & Bagchi, 1962) is able to explain satisfactorily the diffraction effects of quasiperiodic structures, such as natural and synthetic fibres (Kulshreshtha, Patil, Dweltz & Radhakrishnan, 1969; Ochanski, 1967; Schnabel, 1969; Zannetti, Celotti & Fichera, 1969), disordered layer structures like graphite carbons (Ruland, 1965), mixed crystals like Mn spinels (Vogel, 1969) and Fe-Al alloys (Hosemann, Bialas, Schönfeld, Wilke & Weick, 1966); molten metals (Hosemann & Lemm, 1965), vitreous substances like silica and binary glasses (Domenici & Walther, 1965), and liquid crystals (Hosemann, Lemm & Wilke, 1967). All these substances seldom crystallize with a perfect lattice and are, in general, built up from a large number of arbitrarily oriented crystallites of various sizes, each crystallite varying in order. This micro-heterogeneity can be examined by methods of X-ray diffraction line-profile analysis which have been carried out on cold-worked metals and metallic solid solutions (Warren & Averbach, 1952; Wilson, 1962; Langford & Wilson, 1963; Mitra, 1964).

While interpreting the results of line-profile analyses, it should be borne in mind that the lattices in these substances are 'paracrystalline', where the primitive translations of an ideal lattice are replaced by statistically distributed vectors which can vary both in magnitude and direction from cell to cell. These lattice distortions, which destroy the long-range order, are designated as Type II distortions. The diffraction diagrams of paracrystallites show a broadening of the reciprocal lattice node in addition to that caused by the finite size of crystallites. The effects of Type II distortions on the X-ray diffractograms have been studied using an analysis of the optical diffraction patterns of various

lattice models (Bonart, Hosemann & McCullough, 1963; Hosemann & Müller, 1970).

The line-profile studies of this class of substances, especially those involving the calculation of the moments and Fourier transforms of the profile, have rarely been discussed and, in many of the cases where they have, have not been correctly interpreted. Recently, Kulshreshtha, Dweltz & Radhakrishnan (1971*a*) have emphasized the need to distinguish between the diffraction effects of substances having Type I (thermal or frozen-thermal) and Type II (fluid-statistical or paracrystalline) lattice distortions and have contributed to the development of a general theory and a computer-based methodology of line-profile analysis for substances characterized by Type II distortions (Kulshreshtha, Kothari & Dweltz, 1971; Kulshreshtha, Dweltz & Radhakrishnan, 1971*b*).

The present paper describes a theory in which the Fourier transform analysis of the radial diffraction profiles of these substances is used to separate the size and distortion effects. It thus attempts to extend the Warren-Averbach technique of line-profile analysis by Fourier coefficients to the case of a powder of one-dimensional paracrystals by developing the theory to cover Type II distortions. The utility of the expressions presented here is illustrated by an attempt to re-interpret the earlier results of Fourier analysis on linear polyethylene (Katayama, 1961) and petroleum coke powders, heat-treated at various temperatures (Popovic, 1969).

Theory for the Fourier transform analysis of paracrystalline substances

Let us consider a one-dimensional paracrystalline array of lattice points along the \mathbf{b} direction. In Hosemann's (1962) model of an 'ideal paracrystal', the distances between successive lattice points $X_i, i=1, 2, \dots, m, \dots$ constitute a sequence of random variables, which are identically distributed according to the 'coordination statistic' or the probability density $h(t), t=mb$ denoting

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a distance along the b axis. The higher partial statistic $h_m(t)$ is given by the multiple convolution of $h(t)$

$$h_m(t) = \overset{|m|}{\underset{\sim}{h}}(t) \quad (1)$$

and governs the distribution of the distance

$$S_m = X_1 + X_2 + \dots + X_m$$

between the origin and the m th lattice point. The expression on the right hand side of equation (1) means the self-convolution of $h(t)$, $|m|$ - times. Let S'_m denote the displacement of the m th lattice point from its ideal site, relative to the origin. Then

$$\left. \begin{aligned} S'_m &= S_m - \langle S_m \rangle \\ &= S_m - mb \end{aligned} \right\} \quad (2)$$

where $b = \langle X_i \rangle$, is the average spacing between adjacent lattice points and the brackets denote the mathematical expectations of the term they enclose. Variance of S'_m is given by

$$\Delta_m^2 = \langle S_m'^2 \rangle = |m| \Delta^2 \text{ where } \Delta^2 = \langle X_i^2 \rangle. \quad (3)$$

The coefficient of variation of the lattice spacing is

$$g = \frac{\Delta}{b}. \quad (4)$$

The Q -function, *i.e.* the distance-statistics function is given by

$$z(t) = \sum_{m=-\infty}^{\infty} h_m(t) \quad (5)$$

The Fourier transformation of $z(t)$ gives the expression for the interference function, *i.e.* the 'paracrystalline lattice factor' $Z(S)$, where $S = \frac{2 \sin \theta}{\lambda}$ is the distance of

the node $0k0$ from the origin of the reciprocal lattice, and λ the wavelength of X-rays. Thus

$$Z(S) = \sum_{m=-\infty}^{\infty} F.T. [h_m(t)] \quad (6)$$

where

$$F.T. [z(t)] = \int_{-\infty}^{\infty} z(t) \exp(-2\pi i S t) dt$$

denotes the Fourier transform of $z(t)$. The Fourier series representation of $Z(S)$ can be easily understood with the help of equation (6), which gives

$$Z(S) = \sum_{m=-\infty}^{\infty} [Y(k, m) \exp(-2\pi i m b S)], \quad (7a)$$

where the distortion Fourier coefficients are

$$\begin{aligned} Y(k, m) &= J(m) - iK(m) = F.T. [h_m(t + mb)] \\ &= \langle \exp(-2\pi i S S'_m) \rangle = H_m(S) = [H(S)]^{|m|} \end{aligned} \quad (7b)$$

and

$$\begin{aligned} J(m) &= \langle \cos 2\pi S S'_m \rangle = \frac{1}{2} [H_m(S) + H_m^*(S)] \\ K(m) &= \langle \sin 2\pi S S'_m \rangle = -\frac{1}{2i} [H_m(S) - H_m^*(S)]. \end{aligned} \quad (7c)$$

The asterisk (*) denotes the complex conjugate and k the order of reflexion. The meaning of $H_m(S)$ will be discussed in the Appendix. The line profile due to Type II distortions can thus be expressed as

$$Z(s) = \sum_{m=-\infty}^{\infty} [J(m) \cos 2\pi m b s - K(m) \sin 2\pi m b s]$$

where $s = (S - S_0)$ is the actual distance by which S misses the node S_0 of the reciprocal lattice. The Gaussian approximation for $h_m(t + mb)$ will be valid for small $|m|$ if the displacements S'_m are small and also for large $|m|$ by the virtue of the central limit theorem. In this case the imaginary coefficients $K(m)$ will be vanishingly small and the real coefficients (Kulshreshtha, Dweltz & Radhakrishnan, 1971) will be

$$\left. \begin{aligned} J(m) &= \exp(-2\pi^2 \Delta^2 S^2 |m|) \\ \text{or} \\ J(t) &= \exp(-\alpha |t|); \alpha = \frac{2\pi^2 g^2 k^2}{b} \end{aligned} \right\} \quad (8)$$

(The Appendix describes an alternative method of deriving the expressions for the distortion line profile and its Fourier coefficients).

The observed profile, obtained after correction for instrumental factors by the method of Stokes (1948), is a convolution of the size-broadened profile and the distortion-broadened profile. The Stokes corrected real (cos) and imaginary (sin) coefficients are therefore given by

$$\left. \begin{aligned} F_r(t) &= \frac{V(t)}{V(0)} J(t) \\ &= \frac{V(t)}{V(0)} \langle \cos 2\pi S S'_m \rangle \end{aligned} \right\} \quad (9a)$$

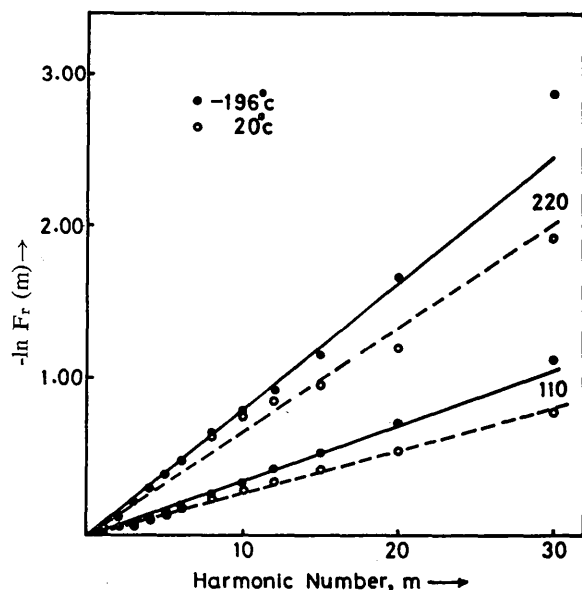


Fig. 1. $-\ln F_r(m)$ vs. m plots for 110 and 220 reflexions of annealed 'Marlex 50' polyethylene filaments measured at -196 and 20°C .

$$\begin{aligned} F_i(t) &= -\frac{V(t)}{V(0)} K(t) \\ &= -\frac{V(t)}{V(0)} \langle \sin 2\pi S S'_m \rangle \end{aligned} \quad (9b)$$

where $V(t)$ is the volume common to the crystal and its ghost obtained through a translation $t=mb$ along the normal to the reflecting $(0k0)$ planes (Wilson, 1962). For small degrees of distortion

$$F_r(t) \simeq \frac{V(t)}{V(0)} \exp(-2\pi^2 S^2 \langle S'_m{}^2 \rangle)$$

or

$$\ln F_r(t) = \ln [V(t)/V(0)] - 2\pi^2 S^2 \langle S'_m{}^2 \rangle \quad (10)$$

Thus, if two orders of reflexion are available, equation (10) can be used to separate the size and distortion effects (Warren & Averbach, 1952). The resulting values $\langle S'_m{}^2 \rangle$ can be interpreted according to equation (3) for Type II distortions and $V(t)/V(0)$ according to standard procedures (Bertaut, 1949; Smith & Simpson, 1965). However, if more than two orders are available, the separation of size and distortion effects is better achieved by Harrison's (1967) method, which yields $V(t)/V(0)$ and various even and odd moments of S'_m as a function of t [*cf.* equations (9a) and (9b)] which, in turn, can be utilized to synthesize the partial statistic $h_m(t+mb)$.

The size coefficients can be approximated as

$$\frac{V(t)}{V(0)} \simeq \exp(-\beta|t|),$$

where

$$\beta = \frac{1}{M_1} = \int \frac{1}{M} g(M) dM \quad (11)$$

and $g(M)$ denotes the frequency function of crystallite lengths M . Expression (11) is a good approximation in the case of fibrous polymers and can be derived in terms of various stochastic models for the placement of crystallites along the length of a fibre. Thus, from equations (8), (9a) and (11):

$$F_r(t) \simeq \exp[-(\alpha + \beta)|t|]$$

or

$$\ln F_r(t) \simeq -\left[\frac{2\pi^2 g^2 k^2}{b} + \frac{1}{M_1} \right] |t|. \quad (12)$$

The utility of this equation will be illustrated with the help of a suitable example. The methodology for computing the Fourier coefficients and thereby obtaining the co-ordination statistic and size distribution of paracrystallites will be discussed elsewhere.

Equilibrium paracrystallite size

Vainshtein (1966) defines the interaction radius t_M of a paracrystalline lattice as the region of order in $z(t)$,

outside which $z(t)$ assumes a constant value, and obtains the following relation

$$t_M = \frac{d_{hkl}}{(2.5g)^2}$$

or

$$g \sqrt{\frac{t_M}{d_{hkl}}} \simeq 0.4. \quad (13)$$

This relation shows that the average size of the coherently diffracting domains is inversely proportional to the degree of disorder g , d_{hkl} denoting the average spacing between the reflecting planes. In practice, however, one obtains the crystallite size values which are much smaller than predicted by equation (13). Hosemann, Loboda-Cackovic & Wilke (1968) define a dimensionless parameter α' as

$$\alpha' = g \sqrt{L/d_{hkl}} \quad (14)$$

where L is the 'weight-average' size of the paracrystallites along the normal to the reflecting planes and is a measure of the micro-heterogeneity of the substance under examination. The use of this parameter is, however, open to criticism since it is obtained through the integral breadth analyses which involve arbitrary assumptions regarding profile shapes. The moment and Fourier transform methods are theoretically sound and very suitable for studying the imperfections in paracrystalline substances and yield the number-average crystallite size M_1 (Guinier, 1963). We therefore define an analogous parameter α'' as

$$\alpha'' = g \sqrt{M_1/d_{hkl}}. \quad (15)$$

The relation between α' and α'' is obviously dependent upon the size distribution of paracrystallites. In general $\alpha' > \alpha''$, and for a Cauchy-like size profile, $\alpha' = \sqrt{2}\alpha''$.

A reinterpretation of earlier results on the Fourier analysis of polyethylene and petroleum cokes

The first study on the Fourier analysis of polymeric X-ray diffraction profiles was carried out by Katayama (1961), who investigated the line broadening in annealed 'Marlex 50' polyethylene filaments at two different temperatures (-196°C and 20°C). He observed that the distortion line profiles had an approximately Cauchy form, but his interpretation of the Fourier coefficients was incorrect, being based on the stress-strain model of disorder. As a result of this erroneous interpretation, he obtained an infinite crystallite size in polyethylene and his estimate of strain was at least an order of magnitude higher than those found in cold-worked metals. Obviously, such a large crystallite size cannot be envisaged in fibres. Later Hosemann, Balta-Calleja & Wilke (1966) could prove by means of integral breadth analysis that lattice distortions in various linear and branched polyethylenes were 'paracrystalline' in character and obtained paracrystallite

Table 1. Condensed results of X-ray diffraction line-profile analysis on paracrystalline aggregates

Reference	Substance	Sample specifications	Lattice planes	d_{hkl}	L	\bar{M}_1	g	α'	α''
Present work, data of Popovic (1969)	Petroleum coke powder	Heat-treated at	$\left\{ \begin{array}{l} 1190^\circ\text{C} \\ 1810 \\ 2050 \\ 2230 \\ 2550 \\ 2850 \end{array} \right\}$	002	3.47 Å	32 Å	4.12%	0.13	
				002	3.43	85	3.28	0.16	
				002	3.43	200	1.83	0.14	
				002	3.38	320	1.64	0.16	
				002	3.37	435	1.14	0.13	
			002	3.36	565	1.02	0.13		
Present work, data of Katayama (1961)	Linear polyethylene	Marlex-50, studied at	$\left\{ \begin{array}{l} -196^\circ\text{C} \\ 20 \end{array} \right\}$	110	4.10	205	2.80	0.22	
				110	4.10	295	2.60	0.20	
Hosemann, Loboda-Cackovic & Wilke (1968)	Linear polyethylene	Hot-stretched 60:1	$\left\{ \begin{array}{l} \text{Component I} \\ \text{Component II} \end{array} \right\}$	110	4.10	91 Å	3.15	0.15	
				110	4.10	186	2.20	0.15	
Hosemann (1967)	Polyethylene single crystals	$\left\{ \begin{array}{l} \text{Hostalen } G \\ \text{Lupolen 6001 } H \end{array} \right\}$		110	4.10	299	1.88	0.16	
				110	4.10	332	2.14	0.19	
Hosemann, Loboda-Cackovic & Wilke (1968)	Polyethylene single crystals	Crystallized at 85°C, 24 hr		110	4.10	530	1.42	0.16	
				200	3.70	337	1.59	0.15	
Buchanan & Miller (1966)	Isotactic polystyrene	$\left\{ \begin{array}{l} 4-120 \\ 4-165 \end{array} \right\}$		110	10.95	131	4.95	0.17	
				110	10.95	162	2.80	0.11	
Kulshreshtha, Patil, Dweltz & Radhakrishnan (1969)	Ramie	Bleached		020	5.15	460	0.80	0.08	
Kulshreshtha, Dweltz & Radhakrishnan (1971a)	Polynosic viscose	Tufcel		020	5.15	125	1.66	0.08	
				020	5.15	90	1.37	0.08	
Hosemann, Loboda-Cackovic & Wilke (1968)	$\left\{ \begin{array}{l} \text{Mn}_x\text{Fe}_{3-x}\text{O}_4 \text{ spinels} \\ \alpha\text{-Fe} + 3\% \text{ Al}_2\text{O}_3 \end{array} \right\}$	$\left\{ \begin{array}{l} x=1.88 \\ \text{Annealed at } 400^\circ\text{C, } 15 \text{ hr} \\ \text{Annealed at } 800^\circ\text{C, } 20 \text{ hr} \end{array} \right\}$		h \bar{h} 0	3.01	1540	0.85	0.19	
				h \bar{h} l	1.24	250	1.20	0.17	
				h00	1.43	200	1.64	0.19	
				h \bar{h} 0	2.03	250	1.02	0.11	
				h \bar{h} h	1.24	500	0.76	0.15	
				h00	1.43	435	1.10	0.19	
				h \bar{h} 0	2.03	540	0.63	0.10	
Hosemann & Lemm (1965)	Lead	223°C above melting point.			10		12.5		

size values in the range 200–500 Å with g -values of 2–3%.

In Fig. 1, $-\ln F_r(m)$ vs. m plots are given for 110 and 220 reflexions of polyethylene studied at -196 and 20°C , as obtained after replotting Katayama's data ($\ln A_n$ vs. n in his notation). The replotting was done by reading individual values from an enlarged photograph of the curves published by him (Katayama, 1961). The present plots (Fig. 1) are straight lines passing through the origin in conformity with equation (12) derived for a fibrous paracrystal. This behaviour suggests the presence of a Gaussian coordination statistic in the paracrystalline lattice of polyethylene, rather than a Cauchy distribution of strains as concluded by Katayama (1961). Analysis of the data in this way suggests the values of 205 and 295 Å for \bar{M}_1 and g -values of 2.8 and 2.6% respectively for the samples studied at -196°C and 20°C .

Popovic (1969) investigated powdered samples of petroleum coke, heat-treated at various temperatures, by carrying out a Fourier analysis of the 00 l reflexions. He, however, used the Warren–Averbach interpretation (1952) of cold-worked metals and alloys for these substances, which possess a distinctly different lattice structure. He plotted the values of $\langle S_m^2 \rangle^{1/2}$ vs. t , [$\langle (\Delta L)_L^2 \rangle^{1/2}$ vs. L in his notation] for various samples obtained from the Fourier analysis [cf. equation (10)]. In this way, he obtained curves which were far from linear. Fig. 2 illustrates $\langle S_m^2 \rangle$ vs. t plots for various samples obtained after replotting the values taken from the original curves of Popovic (1969). These plots are all straight lines passing through the origin, as is to be expected from the relation (3) for an 'ideal paracrystal'. g -values for various heat-treated samples can be obtained by making use of the slopes of these lines and the d_{002} values listed by Popovic (1969) for these samples. The values of \bar{M}_1 , also listed by him (L_c in his notation), would be complementary to the g -values in understanding the fine structure of these samples.

A summary of results of line-profile analyses on paracrystalline aggregates

Table 1 gives a summary of results derived in the present work from earlier existing data and also includes, for the sake of comparison, the results of Buchanan & Miller (1966), Hosemann & Lemm (1965); Hosemann, Bialas, Schönfeld, Wilke & Weick (1966), Hosemann, Lemm & Wilke (1967), Hosemann & Müller (1970), Kulshreshtha, Patil, Dweltz & Radakrishnan (1969) and Kulshreshtha, Dweltz & Radakrishnan (1971*b*) on various paracrystalline substances.

The results on 'Marlex-50' polyethylene filaments indicate that cooling of the specimen from 20 to -196°C generates a higher lattice disorder as evidenced by a decrease in the paracrystallite size and an increase in the g -value. Further, the g and \bar{M}_1 values, calculated by the present method, for these samples are in good

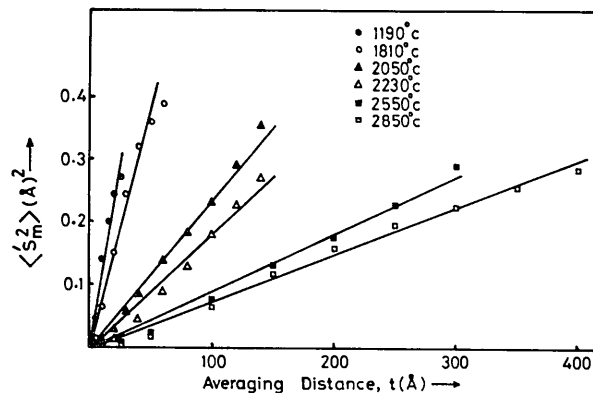


Fig. 2. $\langle S_m^2 \rangle$ vs. t plots for petroleum coke powders heat-treated at various temperatures.

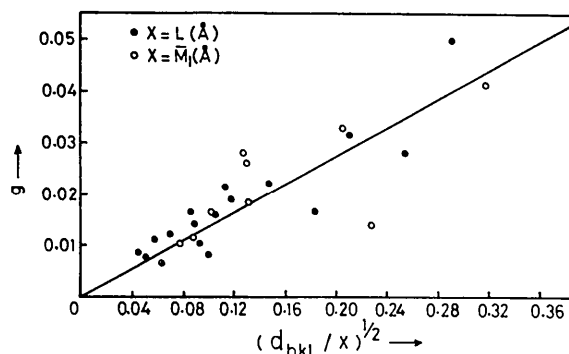


Fig. 3. Correlation between g and $(d_{hkl}/L)^{1/2}$ or $(d_{hkl}/\bar{M}_1)^{1/2}$.

agreement with the values calculated by Hosemann *et al.* (1970) for various polyethylenes.

The results on petroleum cokes show that with increasing temperature of heat-treatment, the lattice order improves as shown by increasing paracrystallite size and decreasing g -values. It can be concluded that the lattice planes or layers in petroleum coke retain a paracrystallite arrangement throughout the process of a gradual transition from the non-graphitic carbons towards the graphitic ones.

Table 1 also lists the values of α' and α'' for various substances. It can be seen that for most of the substances, α' or α'' values are centred around 0.16, whereas for fibres of cellulosic origin they tend to lie around 0.08. A summary of these results is presented in Fig. 3, which shows a plot of the g -value against $(d_{hkl}/\bar{M}_1)^{1/2}$ or $(d_{hkl}/L)^{1/2}$. A correlation coefficient of the order of 0.85 is obtained. The scatter of points, which becomes large as paracrystallite size decreases, may be due to (i) the substances belonging to different classes, (ii) the substances being investigated by different groups of workers and (iii) an inverse inter-relation between the errors in the two parameters.

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APPENDIX

The distance statistics function is

$$z(t) = \delta(t-0) + \sum_{m=1}^{\infty} [h_m(t) + h_{-m}(t)].$$

The lattice factor is

$$\begin{aligned} Z(S) &= F.T. [z(t)] \\ &= 1 + \sum_{m=1}^{\infty} F.T. [h_m(t)] + \sum_{m=1}^{\infty} F.T. [h_m(-t)]. \end{aligned}$$

The Fourier transforms of $h_m(t)$ are called 'statistic amplitudes'. Thus

$$\begin{aligned} F_m(S) &= F.T. [h_m(t)] = H_m(S) \exp(-2\pi i m b S) \\ &= [F(S)]^{|m|} \quad (16) \end{aligned}$$

$$\begin{aligned} F_{-m}(S) &= F.T. [h_m(-t)] = H_m^*(S) \exp(2\pi i m b S) \\ &= [F^*(S)]^{|m|} \end{aligned}$$

where

$$\begin{aligned} H_m(S) &= F.T. [h_m(t + mb)] \\ H_m^*(S) &= F.T. [h_m(-t - mb)] \end{aligned}$$

and * denotes the complex conjugate. We have

$$\begin{aligned} Z(S) &= \sum_{m=0}^{\infty} [F^{|m|} + F^{*|m|}] - 1 \\ &= \frac{1 - |F|^2}{1 + |F|^2 - 2|F| \cos 2\pi b S}, \quad S \neq 0. \quad (17) \end{aligned}$$

For a Gaussian coordination statistic, $|F|$ is given by $\exp(-2\pi^2 g^2 k^2)$ and

$$Z'(S) = \frac{\sinh(2\pi^2 g^2 k^2)}{\cosh(2\pi^2 g^2 k^2) - \cos 2\pi b S}. \quad (18)$$

The line-profile is obtained by replacing S by $s = (S - S_0)$ and becomes, for small g -values

$$Z'(s) \simeq \frac{1}{b} \cdot \frac{2\alpha}{(\alpha^2 + 4\pi^2 s^2)}; \quad \alpha = \frac{2\pi^2 g^2 k^2}{b}. \quad (19)$$

The real distortion Fourier coefficients are

$$\begin{aligned} J(t) &= \int Z'(s) \exp(2\pi i s t) ds \\ &= \exp(-\alpha|t|) \end{aligned}$$

since

$$Z'(s) = \sum_{m=-\infty}^{\infty} \exp(-\alpha b |m| - 2\pi i |m| b s).$$

The coordination statistic is obtainable through the Fourier coefficients $J(m)$ and $K(m)$ [cf. equations (7c) and (16)].

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