

Can Kinematic Diffraction Distinguish Order from Disorder?

Michael Baake*

Fakultät für Mathematik, Universität Bielefeld, Postfach 100131, 33501 Bielefeld, Germany

Uwe Grimm†

*Department of Mathematics and Statistics, The Open University,
Walton Hall, Milton Keynes MK7 6AA, United Kingdom*

(Dated: 30 October 2008)

Diffraction methods are at the heart of structure determination of solids. While Bragg-like scattering (pure point diffraction) is a characteristic feature of crystals and quasicrystals, it is not straightforward to interpret continuous diffraction intensities, which are generally linked to the presence of disorder. However, based on simple model systems, we demonstrate that it may be impossible to draw conclusions on the degree of order in the system from its diffraction image. In particular, we construct a family of one-dimensional binary systems which cover the entire entropy range but still share the same purely diffuse diffraction spectrum.

PACS numbers: 61.05.cc 61.43.-j 61.44.-n 02.50.-r

The inverse problem of reconstructing a structure from its diffraction pattern is one of the most important challenges in materials science. Its degree of complexity increases if one goes beyond simple periodic systems to cover quasicrystals, modulated structures or complex alloys. In particular, it has been realised that the Bragg diffraction alone is generally insufficient for a complete reconstruction^{1,2,3}.

Currently, an increasing effort is being made to understand and utilise the continuous part of the diffraction pattern; see Refs. 1,2 for background and Refs. 4,5 for recent applications. However, even in the idealised situation of a perfect diffraction experiment with unlimited resolution, the reconstruction is generally not unique. Already in 1944, Patterson⁶ discussed *homometric* point sets, which are point sets whose kinematic diffraction patterns coincide, and provided explicit examples to illustrate the ambiguity. It was demonstrated³ that it may be possible to lift the ambiguity, and thus to determine the structure uniquely, if higher-order correlations are known. While one can argue that, for structures originating from systems with pure pair-potential interaction (or allowing a description by effective pair potentials, compare Ref. 7), higher-order correlations are determined by the pair correlations⁸, this is not generally the case, and in practice measurements of higher-order correlations are extremely difficult. The role of phase information in stochastic systems was investigated in Ref. 9.

Here, we want to go one step further, and compare the diffraction of various point sets, ranging from deterministic to fully stochastic, in a parametrised way. We characterise the degree of order by the corresponding (metric) entropy. As we will see below, it is possible to construct families of point sets which span an entire entropy range but share the same kinematic diffraction pattern – proving that diffraction is insensitive even to the degree of order in this case.

We start by giving a brief introduction to some basic notions of mathematical diffraction theory, for a one-

dimensional (but relevant) setting with scatterers placed on integer positions. Although this is a highly idealised situation that ignores displacement effects, its practical relevance is well known; see Ref. 1 and references therein. Then, we discuss the diffraction of two binary systems (characterised by two scattering strengths) – a perfectly ordered structure based on a specific deterministic sequence, and a completely random structure based on a coin-tossing experiment. It was observed earlier¹⁰ that these rather different systems share the same diffraction, and are thus homometric. Finally, we introduce a ‘Bernoullisation’ procedure to couple coin-tossing disorder to a perfectly ordered structure, thus producing partially ordered systems of varying entropy. We employ this procedure to explicitly construct a family of binary systems which are homometric and cover the entire available entropy range from the perfectly ordered (entropy 0) to the fully stochastic situation (entropy $\log(2)$). Although these systems may not occur naturally, they can be made synthetically. In the simplest scenario, a binary structure can be produced by sequential deposition of layers consisting of two different materials, which makes it possible to realise any desired sequence; see Ref. 11 for an example. More complicated structures are also feasible, see for instance Ref. 12, and such artificial materials with designed physical properties will become increasingly important.

a. Diffraction of Dirac combs. To keep arguments simple, we consider the diffraction of one-dimensional systems with point-like scatterers located at integer points $n \in \mathbb{Z}$. The scattering strengths are given by weights w_n for $n \in \mathbb{Z}$, which we assume to be real for simplicity (the setting can be extended to complex weights). The corresponding scattering density is modelled by the *Dirac comb*

$$\omega = \sum_{n \in \mathbb{Z}} w_n \delta_n,$$

where δ_x denotes the normalised point measure (Dirac δ) on the real line, located at position x . Clearly, all distances between scatterers are integer valued. This implies that the *autocorrelation* (or Patterson) measure γ , assuming its existence for the moment, is again a Dirac comb on \mathbb{Z} ,

$$\gamma = \sum_{m \in \mathbb{Z}} \eta(m) \delta_m, \quad (1)$$

with the coefficients $\eta(m)$ obtained as the limits

$$\eta(m) = \lim_{N \rightarrow \infty} \frac{1}{2N+1} \sum_{n=-N}^N w_n w_{n+m}.$$

The scattering intensity $I(k)$ for wave numbers $k \in \mathbb{R}$ is then determined by the *diffraction* measure $\hat{\gamma}$, the Fourier transform of the autocorrelation γ , compare Ref. 13 for background. There are several slightly different versions of the Fourier transform. We prefer to use

$$\hat{\phi}(k) = \int_{\mathbb{R}} e^{-2\pi i k x} \phi(x) dx$$

for a Schwartz function ϕ , and its standard extension to tempered distributions and measures; see Ref. 14 for details.

For the case of a one-dimensional crystal with $w_n = w$ for all $n \in \mathbb{Z}$, we have $\eta(m) = w^2$ for all $m \in \mathbb{Z}$, hence $\gamma = w^2 \delta_{\mathbb{Z}}$, where we use $\delta_{\mathbb{Z}}$ as shorthand for the sum $\sum_{n \in \mathbb{Z}} \delta_n$. Its Fourier transform is obtained by Poisson's summation formula¹⁴, $\hat{\delta}_{\mathbb{Z}} = \delta_{\mathbb{Z}}$, which gives $\hat{\gamma} = w^2 \delta_{\mathbb{Z}}$. The diffraction image thus consists entirely of Bragg peaks, located at integer positions k , with equal diffraction intensities $I(k) = w^2$. The diffraction spectrum in this case is *pure point*, meaning that it consists of Bragg peaks only. In general, the diffraction measure may comprise three different contributions,

$$\hat{\gamma} = \hat{\gamma}_{\text{pp}} + \hat{\gamma}_{\text{sc}} + \hat{\gamma}_{\text{ac}}$$

where $\hat{\gamma}_{\text{pp}}$ is the pure point part, consisting of a countable sum of δ peaks. The term $\hat{\gamma}_{\text{ac}}$ corresponds to the *absolutely continuous* component, which can be described by a locally integrable (and often continuous) non-negative function $I_{\text{ac}}(k)$ of the wave vector k . The remainder, if there is any, is called the *singular continuous* component $\hat{\gamma}_{\text{sc}}$. While it vanishes on the complement of a set S of measure 0, even within S it never gives weight to any single point. When such a component is present, S can thus not be a countable set. Apart from trivial examples of a diffraction measure that is concentrated on a line in the plane, or similarly on a manifold of lower dimension, typical examples for this strange contribution are diffraction intensities which are supported on a Cantor set or a dense set. A well-known example for the latter phenomenon is the Thue-Morse chain. For an appropriate choice of its scattering strengths, it has a purely singular continuous diffraction measure; see Refs. 15,16,17 for derivations and Refs. 11,12 for applications.

If the Dirac comb on \mathbb{Z} is *periodic*, which means that there is an integer $p > 0$ such that $w_{n+p} = w_n$ for all $n \in \mathbb{Z}$, the diffraction measure is pure point, and supported on the lattice \mathbb{Z}/p . It is again periodic, at least with period 1, but not necessarily with any smaller period. As an example, consider the alternating Dirac comb with $w_n = (-1)^n$. In this case, $\eta(m) = (-1)^m$ for $m \in \mathbb{Z}$, so the autocorrelation is $\gamma = \delta_{2\mathbb{Z}} - \delta_{2\mathbb{Z}+1}$. By Poisson's summation formula and elementary properties of the Fourier transform (such as the behaviour under scaling and the convolution theorem), we have

$$\widehat{\delta_{2\mathbb{Z}+1}} = \frac{\cos(2\pi k)}{2} \delta_{\mathbb{Z}/2},$$

which leads to the diffraction measure

$$\hat{\gamma} = \frac{1 - \cos(2\pi k)}{2} \delta_{\mathbb{Z}/2} = \delta_{\mathbb{Z}+1/2}.$$

In this case, the diffraction spectrum is again pure point, and consists of Bragg peaks of unit intensity at positions $n + 1/2$, hence on a subset of $\mathbb{Z}/2$. The fundamental period of $\hat{\gamma}$ is nevertheless still 1.

To obtain absolutely or singular continuous components, in line with the classification of Ref. 18, we thus have to go beyond the periodic situation.

b. Rudin-Shapiro versus Bernoulli. Let us start with a deterministic system without periodicity, based on the well-known binary Rudin-Shapiro chain. We consider the corresponding Dirac comb

$$\omega_{\text{RS}} = \sum_{n \in \mathbb{Z}} w(n) \delta_n,$$

where $w: \mathbb{Z} \rightarrow \{\pm 1\}$ is defined by the recursion

$$w(4n + \ell) = \begin{cases} w(n), & \text{for } \ell \in \{0, 1\}, \\ (-1)^{n+\ell} w(n), & \text{for } \ell \in \{2, 3\}, \end{cases} \quad (2)$$

together with the two initial conditions $w(0) = 1$ and $w(-1) = -1$. The resulting system is an aperiodic sequence in 1 and -1 , both appearing equally frequent; see Fig. 1 for a graphical representation. It has many nice properties, such as strict ergodicity and linear patch counting complexity, see Ref. 16 and references therein for details. In particular, these properties imply that this sequence has topological (and metric) entropy 0.

Since ω_{RS} is a Dirac comb on \mathbb{Z} , the autocorrelation is of the form (1), with coefficients

$$\eta_{\text{RS}}(m) = \lim_{N \rightarrow \infty} \frac{1}{2N+1} \sum_{n=-N}^N w(n) w(n+m).$$

It follows from the unique ergodicity of the RS sequence that all these coefficients (and hence γ) exist, by an application of the ergodic theorem. They are given by $\eta_{\text{RS}}(m) = \delta_{m,0}$, which is astonishing. The construction of a deterministic sequence with vanishing two-point correlations was the original (and independent) motivation



FIG. 1: Central part of the Rudin-Shapiro chain, with open (full) circles representing scattering strengths $w(n) = 1$ ($w(n) = -1$), respectively. The location of the origin ($n = 0$) is indicated by the vertical line.

of Rudin and Shapiro, thus answering a question in the theory of Fourier series.

Let us prove this property by a simple, explicit argument. Consider $a_m = \eta_{\text{RS}}(m)$ together with

$$b_m = \lim_{N \rightarrow \infty} \frac{1}{2N+1} \sum_{n=-N}^N (-1)^n w(n) w(n+m),$$

which also exist (by another application of the ergodic theorem). Clearly, one has $a_0 = 1$ and $b_0 = 0$, because $w(n)^2 = 1$ for all $n \in \mathbb{Z}$. Then, considering m modulo 4, and splitting the sums in the definition of a_m and b_m accordingly, the recursion relations (2) imply

$$\begin{aligned} a_{4m} &= \frac{1+(-1)^m}{2} a_m, & a_{4m+2} &= 0, \\ a_{4m+1} &= \frac{1-(-1)^m}{4} a_m + \frac{(-1)^m}{4} b_m - \frac{1}{4} b_{m+1} \\ a_{4m+3} &= \frac{1-(-1)^m}{4} a_{m+1} - \frac{(-1)^m}{4} b_m + \frac{1}{4} b_{m+1}. \end{aligned}$$

Similarly, one finds

$$\begin{aligned} b_{4m} &= 0, & b_{4m+2} &= \frac{(-1)^m}{2} (b_m + b_{m+1}), \\ b_{4m+1} &= \frac{1-(-1)^m}{4} a_m - \frac{(-1)^m}{4} b_m + \frac{1}{4} b_{m+1}, \\ b_{4m+3} &= -\frac{1+(-1)^m}{4} a_{m+1} - \frac{(-1)^m}{4} b_m + \frac{1}{4} b_{m+1}. \end{aligned}$$

Using the initial data, these recursion relations imply that $a_m = b_m = 0$ for all integers $m \neq 0$. This result shows that the autocorrelation and diffraction measures of the binary Rudin-Shapiro Dirac comb ω_{RS} are simply

$$\gamma_{\text{RS}} = \delta_0 \quad \text{and} \quad \widehat{\gamma}_{\text{RS}} = \lambda,$$

where λ denotes Lebesgue measure. In other words, the diffraction measure is purely absolutely continuous, and consists of a constant background (of height 1) only. The extinction of all Bragg peaks is due to the balanced choice of weights. This is convenient for the theoretical argument, but also relevant in practice¹⁹ when disregarding thermal displacement. Note that all arguments can be extended to mixed spectra.

Perhaps the most elementary stochastic system is based upon the classic coin-tossing (or Bernoulli) experiment. We consider a stochastic Dirac comb²⁰ on \mathbb{Z} ,

$$\omega_{\text{B}} = \sum_{n \in \mathbb{Z}} W_n \delta_n,$$

where $(W_n)_{n \in \mathbb{Z}}$ is a family of independent identically distributed (i.i.d.) random variables, with probabilities $\mathbb{P}(W_n = 1) = p$ and $\mathbb{P}(W_n = -1) = 1 - p$, where $0 \leq p \leq 1$. The corresponding (metric) entropy is

$$H(p) = -p \log(p) - (1-p) \log(1-p), \quad (3)$$

which satisfies $0 \leq H(p) \leq \log(2)$. It attains the extremal values for $p = 0$ and $p = 1$, where $H = 0$ is minimal, and for $p = \frac{1}{2}$, where $H = \log(2)$ is maximal.

The autocorrelation $\gamma_{\text{B}} = \sum_{n \in \mathbb{Z}} \eta_{\text{B}}(m) \delta_m$ is once again a pure point measure that is supported on \mathbb{Z} , with autocorrelation coefficients given by

$$\eta_{\text{B}}(m) = \lim_{N \rightarrow \infty} \frac{1}{2N+1} \sum_{n=-N}^N W_n W_{n+m} \quad (4)$$

for $m \in \mathbb{Z}$. These coefficients almost surely exist, by an application of the strong law of large numbers (see below), for all $m \in \mathbb{Z}$, and satisfy

$$\eta_{\text{B}}(m) = \begin{cases} 1, & m = 0, \\ (2p-1)^2, & m \neq 0. \end{cases}$$

This statement can be proved as follows. It obviously holds for $m = 0$, so consider some fixed $m \neq 0$. The products $Z_n := W_n W_{n+m}$ form a family $(Z_n)_{n \in \mathbb{Z}}$ of identically distributed random variables, which take the values 1 and -1 with probabilities $p^2 + (1-p)^2$ and $2p(1-p)$, respectively. These new random variables are not independent, but we can split the sum in Eq. (4) into two sums (for instance according to even and odd values of $\lfloor \frac{n}{m} \rfloor$, the largest integer smaller than or equal to $\frac{n}{m}$). The resulting two sums each comprise pairwise independent random variables. An application of the strong law of large numbers in its formulation by Etemadi²¹ then shows that each sum almost surely converges (as $N \rightarrow \infty$) to the expectation value of any of the single random variables in the sum, which is $\frac{1}{2}(2p-1)^2$. Hence, the diffraction measure of the stochastic Dirac comb ω_{B} almost surely is

$$\widehat{\gamma}_{\omega_{\text{B}}} = (2p-1)^2 \delta_{\mathbb{Z}} + 4p(1-p) \lambda,$$

where λ again denotes Lebesgue measure. In other words, the diffraction spectrum comprises a constant background of intensity $4p(1-p)$ for any value of the wave number k and Bragg peaks of intensity $(2p-1)^2$ at integer k . Note that, for the perfectly ordered cases $p = 0$ and $p = 1$, the background vanishes, while the Bragg peaks vanish for the maximally disordered case $p = \frac{1}{2}$. At this value of p , the diffraction measure coincides with that of the Rudin-Shapiro chain.

This establishes the homometry of the deterministic binary Rudin-Shapiro chain (with entropy 0) and the completely random Bernoulli chain with $p = \frac{1}{2}$ (with entropy $\log(2)$), as originally observed in Ref. 10. Coupling the two systems in a suitable way, we now extend this to an entire family that covers the intermediate entropy range.

c. Bernoullisation. The Bernoulli chain discussed above is an example of a completely random and interaction-free system. In view of real world examples, it is interesting to explore what happens if one imposes the influence of coin tossing on the order of a deterministic system. This can be realised in many different ways. Here, we focus on binary sequences and modify them by an i.i.d. family of Bernoulli variables.

Consider a bi-infinite binary sequence $S \in \{\pm 1\}^{\mathbb{Z}}$ which we assume to be uniquely ergodic. Then, the corresponding Dirac comb $\omega_S = \sum_{n \in \mathbb{Z}} S_n \delta_n$ possesses the unique (natural) autocorrelation $\gamma_S = \sum_{m \in \mathbb{Z}} \eta_S(m) \delta_m$ with the autocorrelation coefficients $\eta_S(m)$, where $\eta_S(0) = 1$ by construction.

Let $(W_n)_{n \in \mathbb{Z}}$ be an i.i.d. family of random variables that each take values $+1$ and -1 with probabilities p and $1 - p$. The ‘Bernoullisation’ of ω_S is the random Dirac comb

$$\omega := \sum_{n \in \mathbb{Z}} S_n W_n \delta_n, \quad (5)$$

which emerges from ω_S by independently changing the sign of each S_n with probability $1 - p$. Setting $Z_n := S_n W_n$ defines a new family of independent (though in general not identically distributed) random variables, with values in $\{\pm 1\}$. Despite this modification, the autocorrelation γ of ω almost surely exists and can be determined via its autocorrelation coefficients $\eta(m)$ as follows. Since one always has $\eta(0) = \eta_S(0) = 1$, let $m \neq 0$ and consider, for large N , the sum

$$\begin{aligned} \frac{1}{2N+1} \sum_{n=-N}^N Z_n Z_{n+m} &= \\ \frac{1}{2N+1} \left(\sum_{(+,+)} + \sum_{(-,-)} - \sum_{(+,-)} - \sum_{(-,+)} \right) W_n W_{n+m}, \end{aligned}$$

which is split according to the value of (S_n, S_{n+m}) . Each of the four sums can then be handled in the same way as in the argument for the Bernoulli chain above, thus contributing $(2p - 1)^2$ times the frequency of the corresponding sign pair. Observing that the overall signs are the products $S_n S_{n+m}$, it is clear that, as $N \rightarrow \infty$, one (almost surely) obtains

$$\eta(m) = (2p - 1)^2 \eta_S(m)$$

for all $m \neq 0$. This shows that the new autocorrelation almost surely is

$$\gamma = (2p - 1)^2 \gamma_S + 4p(1 - p) \delta_0$$

where γ_S is the unique autocorrelation of ω_S .

Let us apply this Bernoullisation procedure to the Rudin-Shapiro chain. Denote by ω the random Dirac comb obtained from the Bernoullisation (with parameter p) of the binary Rudin-Shapiro chain. Then, the autocorrelation measure almost surely exists and reads $\gamma = \delta_0$,

independently of p . This means that the random Dirac combs ω , even for different values of p , are almost surely homometric, and share the purely absolutely continuous diffraction measure $\hat{\gamma} = \lambda$.

Note that this example explores the full entropy range of Eq. (3): the Bernoulli case (with $p = \frac{1}{2}$) has entropy $\log(2)$, the maximal value for a binary system, while Rudin-Shapiro has entropy 0, and the parameter p interpolates continuously between the two limiting cases. The solution of the corresponding inverse problem is thus highly degenerate. Unless additional information is available, for instance via higher order correlations, one possible strategy could employ a maximum entropy method²², singling out the Bernoulli comb.

Both the Bernoullisation procedure and the specific one-dimensional examples immediately generalise to higher dimensions by taking direct product structures. In particular, the product of d Rudin-Shapiro chains results in a deterministic system in d -space with the same purely absolutely continuous diffraction measure as the corresponding coin-tossing model. Consequently, our above conclusions extend to this case. This means that one can also produce examples with lower rank entropy, which is a new phenomenon in dimensions $d \geq 2$.

d. Concluding remarks. Diffraction methods provide the most important approach to structure determination. The presence of Bragg diffraction clearly indicates an ordered structure, though the discovery of quasicrystals²³ in the 1980s has shown that pure point diffraction occurs in more general systems than just conventional crystals. To date, the precise atomic structure of quasicrystalline alloys is still not completely understood; there is evidence that entropy plays an important role in stabilising quasicrystalline structures, and that some disorder may be an inherent feature of these alloys. Like for many ordinary crystals, diffuse scattering is present in experimental diffraction patterns of even the best known quasicrystals, and there is an increasing effort to explore the information contained the diffuse diffraction intensity^{1,2}; see Ref. 4 for a recent example. It is tempting to draw conclusions about the degree of order in a structure on the basis of the observed diffuse scattering intensity. However, as our explicit example demonstrates, such conclusions have to be considered carefully, since the relation between diffuse scattering and disorder is far from simple.

Acknowledgments

This work was supported by the German Research Council (DFG), within the CRC 701, and by EPSRC via Grant EP/D058465. It is a pleasure to thank the School of Mathematics and Physics at the University of Tasmania for their kind hospitality.

-
- * mbaake@math.uni-bielefeld.de
- † u.g.grimm@open.ac.uk
- ¹ T. R. Welberry, *Diffuse X-Ray Scattering and Models of Disorder*, vol. 6 of *IUCr Monographs on Crystallography* (Oxford University Press, Oxford, 2004).
 - ² R. B. Neder and T. Proffen, *Diffuse Scattering and Defect Structure Simulations*, vol. 11 of *IUCr Texts on Crystallography* (Oxford University Press, Oxford, 2008).
 - ³ F. A. Grünbaum and C. C. Moore, *The use of higher-order invariants in the determination of generalized Patterson cyclotomic sets*, *Acta Cryst. A* **51**, 310–323 (1995).
 - ⁴ M. de Boissieu, S. Francoual, Y. Kaneko, and T. Ishimasa, *Diffuse scattering and phason fluctuations in the Zn-Mg-Sc icosahedral quasicrystal and its Zn-Sc periodic approximant*, *Phys. Rev. Lett.* **95**, 105503 (2005).
 - ⁵ M. Kopecký, J. Fábry, J. Kub, A. Lausi, and E. Busetto, *Multi-energy anomalous diffuse scattering*, *Phys. Rev. Lett.* **100**, 195504 (2008).
 - ⁶ A. L. Patterson, *Ambiguities in the X-ray analysis of crystal structures*, *Phys. Rev.* **65**, 195–201 (1944).
 - ⁷ W. Schweika and A. E. Carlsson, *Short-range order in Ising-like models with many-body interactions: Description via effective pair interactions*, *Phys. Rev. B* **40**, 4990–4999 (1989).
 - ⁸ D. M. C. Nicholson, R. I. Barabash, G. E. Ice, C. J. Sparks, J. L. Robertson, and C. Wolverton, *Relationship between pair and higher-order correlations in solid solutions and other Ising systems*, *J. Phys.: Condens. Matter* **18**, 11585–11594 (2006).
 - ⁹ T. R. Welberry and R. L. Withers, *The role of phase in diffuse diffraction patterns and its effect on real-space structure*, *J. Appl. Cryst.* **24**, 18–29 (1991).
 - ¹⁰ M. Höffe and M. Baake, *Surprises in diffuse scattering*, *Z. Kristallogr.* **215**, 441–444 (2000).
 - ¹¹ F. Axel and H. Terauchi, *High-resolution x-ray-diffraction spectra of Thue-Morse GaAs-AlAs heterostructures: Towards a novel description of disorder*, *Phys. Rev. Lett.* **66**, 2223–2226 (1991).
 - ¹² W. Steurer and D. Sutter-Widmer, *Photonic and phononic quasicrystals*, *J. Phys. D: Appl. Phys.* **40**, R229–R247 (2007).
 - ¹³ A. Hof, *On diffraction by aperiodic structures*, *Commun. Math. Phys.* **169**, 25–43 (1995).
 - ¹⁴ L. Schwartz, *Théorie des Distributions* (Herman, Paris, 1998), 3rd ed.
 - ¹⁵ S. Kakutani, *Strictly ergodic symbolic dynamical systems*, in *Proceedings of the Sixth Berkeley Symposium on Mathematical Statistics and Probability*, edited by L. M. Le Cam, J. Neyman, and E. L. Scott (University of California Press, Berkeley, 1972), pp. 319–326.
 - ¹⁶ M. Queffélec, *Substitution Dynamical Systems – Spectral Analysis*, vol. 1294 of *Lecture Notes in Mathematics* (Springer, Berlin, 1987).
 - ¹⁷ M. Baake and U. Grimm, *The singular continuous diffraction measure of the Thue-Morse chain*, *J. Phys. A: Math. Theor.* **41**, 422001 (2008).
 - ¹⁸ M. Baake and R. V. Moody, *Dirac combs with pure point diffraction*, *J. reine angew. Math. (Crelle)* **573**, 61–94 (2004).
 - ¹⁹ S. Dietrich and W. Fenzl, *Diffuse scattering of x rays or neutrons from binary alloys and null matrices*, *Phys. Rev. B* **39**, 8900–8906 (1989).
 - ²⁰ M. Baake and R. V. Moody, *Diffraction point sets with entropy*, *J. Phys. A: Math. Gen.* **31**, 9023–9039 (1998).
 - ²¹ N. Etemadi, *An elementary proof of the strong law of large numbers*, *Z. Wahrscheinlichkeitsth. verw. Gebiete* **55**, 119–122 (1981).
 - ²² S. Guiaşu and A. Shenitzer, *The principle of maximum entropy*, *Math. Intelligencer* **7**, 42–48 (1985).
 - ²³ D. Shechtman, I. Blech, D. Gratias, and J. W. Cahn, *Metallic phase with long-range orientational order and no translational symmetry*, *Phys. Rev. Lett.* **53**, 1951–1953 (1984).