Powder diffraction from a combinatorial and analytic viewpoint

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Abstract

The mathematical theory of the powder diffraction intensity function is presented in the form accessible to the theoretical crystallography audience. The theory elucidates how a-priori estimates and values of certain averages, or moments of the intensity function bear witness to the fractal dimension and symmetry structure of the material under investigation.

While peak analysis is today's key method of processing the diffraction data, this paper stresses the importance of the moments of the intensity function. The moments are easy to compute and are robust to noise and errors. On the other hand, they represent a unique signature of a particular underlying symmetry type and generally tend to increase with the extent of the order in the material's crystal structure. Supposedly, the moments reach a maximum in the case of the cubic lattice, and this is closely related to the Erdös distance conjecture in combinatorics and analysis which has been open for almost sixty years.

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1 Introduction

Powder diffraction is one of the most widely used material characterization methods. Over the last 50 years it has been routinely used for crystalline phase "finger-printing". See, for example, [17], and the reference contained therein. Recently, cheap and powerful computers, and dedicated 2nd and 3rd generation X-ray synchrotron sources have transformed powder diffraction into a very potent structural tool.

The principal limitation of powder diffraction is the fact that a three-dimensional set of spots obtained from a single crystal experiment is condensed into one dimension. This leads to both accidental and exact peak overlaps, and complicates the determination of exact peak intensities. As a consequence, crystal symmetry cannot be seen directly from diffraction pattern. See, for example, [16] and [1] for the discussion of the errors involved.

The purpose of this article is to present to the audience of theoretical crystallographers the state-of-the art of the theory of the n-dimensional[1] powder diffraction intensity function. The latter is a real-valued non-negative function of one non-negative real variable. Over the past twenty years, it has been an important object of intense mathematical study in the context of distance sets theory in geometric combinatorics and measure theory. In many cases, mathematicians who conducted these investigations did not even suspect that the object of their analysis has such interesting and profound physical significance. Below, we review and conceptualize several important estimates of the intensity function, some with and others without proofs. This paper will introduce the reader to the extensive body of mathematical literature on the subject, and some most recent results are presented here for the first time.

Our basic object of study is a sample, represented by a set $E \subset \mathbb{R}^n$. The length scale is chosen so that E lies in the unit cube $[0,1]^n$, and the diameter of E is also approximately one. First we will consider the case when E is a discrete set, and we later move on to the continuous case. In both situations, μ_E is a normalized measure supported on E. In the discrete case, μ_E is the sum of δ-functions, normalized by the number of elements in E. In the continuous case it is typically a density function, whose integral over E equals one. The *structure function* of E is represented by the Fourier transform

$$F_E(\boldsymbol{\xi}) = \int e^{-2\pi i \boldsymbol{\xi}\cdot\boldsymbol{x}} d\mu_E(\boldsymbol{x}). \tag{1.1}$$

Since only the absolute value of $F_E(\boldsymbol{\xi})$ is observable in a diffraction experiment, define

$$I_E(\boldsymbol{\xi}) = |F_E(\boldsymbol{\xi})|^2, \tag{1.2}$$

a non-negative real-valued function to be the intensity function. Our objective is to show how the structure of E is encoded into certain averages of the intensity function. In particular, within

[1] We keep n as a parameter to emphasize that the results presented are not confined to the case $n = 3$. Moreover, from the mathematical point of view, the most difficult dimension is $n = 2$, while the case $n > 4$ may be easier. See the discussion in the main body of the paper.

a class of sets E under consideration, which ones maximize the value of the intensity function $I_E(\boldsymbol{\xi})$ in a suitable sense?

We shall primary study the *spherical average*

$$\sigma_E(r) = \int I_E(\boldsymbol{\xi}) d\omega_{\boldsymbol{\xi}} \tag{1.3}$$

where $d\omega_{\boldsymbol{\xi}}$ is the surface area measure on the unit sphere S^{n-1} in the reciprocal space.

Of course, in real powder diffraction experiments, the variable is the angle θ inside the cone of X-ray radiation, scattered by the crystalline powder. The radiation has fixed wavelength λ, which is equivalent to fixing $\|\boldsymbol{\xi}\|$ in (1.3). However, from the mathematical point of view, in order to study the direction average which occurs during powder diffraction, it is important to allow λ, or rather its surrogate $\|\boldsymbol{\xi}\|$ to vary. Indeed, the variables λ and θ relate to each other via the Bragg formula.

Spherical averages of the squares of the Fourier transforms of measures constitute the core of harmonic analysis, and have numerous applications in number theory and related disciplines. See for example, [15], [13], and the references contained therein. The quantity $\sigma_E(r)$ is also the main feature of this article.

Observe that the spherical average is interesting only for $r > 1$, as I_E equals 1 at the origin (the integral $\int d\mu_E$) and approximately 1 for small r, by the uncertainty principle.

If the set E is suitably random, the spherical average $\sigma_E(r)$ is quite regular in the sense that it tends to 0 as $r^{-\beta}$ for some $\beta \in (0, n)$ which depends on the dimension of E. On the flip side, if the set E possesses a lattice structure, the quantity $\sigma_E(r)$ displays peaks, which are usually narrow and well separated from one another to ensure that on average over large intervals of r, the spherical average still has mean decay $\sim r^{-\beta}$. However the point-wise estimate $\sigma_E(r) \leq \text{const.}r^{-\beta}$ for all $r > 1$ is no longer valid.

This underlies the peak analysis, routinely performed by the practitioners of powder diffraction in order to document or identify different crystalline phases. Namely, the quantity $\sigma_E(r)$ is analyzed point-wise. From the mathematical point of view, point-wise analysis of functions is most difficult, and is often eschewed in favor of certain averages, or moments. If we extend the function $\sigma_E(r)$ to the whole space $\mathbb{R}^n$ by viewing it as a radially symmetric function of $\boldsymbol{\xi} \in \mathbb{R}^n$, then let for $l = 1, 2, \ldots$

$$M_{E,2l} = \int \sigma_E^{2l}(\boldsymbol{\xi}) d\boldsymbol{\xi} = \int_0^\infty \sigma_E^{2l}(r) r^{n-1} dr \tag{1.4}$$

if the integrals converge. If they do not, we will restrict integration to the interval of length $R \gg 1$ in the last integral and study the dependence of the result on the parameter R.

We will show that the *second moment* $M_{E,2}$ (corresponding to $l = 1$) represents a very important characteristic of the *distance set* of E, defined as

$$\Delta(E) = \{\|\boldsymbol{x} - \boldsymbol{y}\| : \boldsymbol{x}, \boldsymbol{y} \in E\}, \tag{1.5}$$

where $\|\cdot\|$ denotes the Euclidean distance. In particular, the greater $M_{E,2}$, the smaller the distance set $\Delta(E)$ may be. In the discrete case, if E is a "typical" set of N elements, one can expect to have some $N(N-1)/2$ distinct distances in the distance set $\Delta(E)$. However, if E is translation-invariant, the distance set gets considerably smaller. It is conjectured that of all the discrete sets of N elements (clearly modulo rigid motions and scalings), the minimum number of distances occurs in the case of the $\sqrt[n]{N} \times \ldots \sqrt[n]{N}$ integer grid.

The fourth and higher moments of the spherical average also turn out to to be indicative of translation-invariant structure, and perhaps in a more explicit way than the second moment. To this effect, we shall show that if the $2l$th, for $l = 2, 3, \ldots$ moment is large enough, then the set belongs to a finite union of lattices. As a counterpart to this result, we shall show that sets with no translation-invariant structure have small fourth moments.

The main theme of this paper is that averages of the intensity function alluded to above are still sensitive to the fundamental geometric properties of the sample set E and should be useful for material fingerprinting and related purposes whenever powder diffraction is involved. While they contain less general information than the whole function $\sigma_E(r)$, they are easier to compute, store and compare, and, being averages, are relatively insensitive to noise and other errors.

This paper is organized as follows. The first section deals with the discrete case, with allusions to the Erdös distance conjecture, which has been a major unsolved problem in geometrical combinatorics since 1946. In Section 3 we will study the continuous case, in particular the case when the set E can be characterized by some fractal dimension $\alpha \in (0, n)$. Throughout the discussion we emphasize the synthesis of the ideas we present, and prospects for further mathematical development of this rich and important area of modern physics.

2 Discrete case

In this section we operate on the length scale where an average distance between the atoms in the sample studied is approximately one.

We shall make use of the following terminology.

Definition 2.1. An infinite discrete set $A \subset \mathbb{R}^n$ is a *Delaunay* set if

i. there exists a constant $c_A > 0$ such that $\|\boldsymbol{a} - \boldsymbol{a}'\| \geq c_A$ for every pair of different elements $\boldsymbol{a}, \boldsymbol{a}' \in A$,

ii. there exists a constant $C_A > c_A$ such that every cube of side-length C_A contains at least one element of A.

Without loss of generality we may assume that a Delaunay set A contains exactly one point in each cube of the form

$$[0,1]^n + m, \ m \in \mathbb{Z}^n.$$

We shall assume in the sequel that A is of this form.

Let $q \gg 1$ and $A_q = A \cap [0,q]^n$ be a truncation of A. I.e. the sample set E is $\frac{1}{q}A_q$. In view of the general discussion in the introduction, the corresponding measure, normalized by the number of elements is

$$\mu_{A_q} = \frac{1}{q^n} \sum_{\boldsymbol{a} \in A} \delta(\boldsymbol{x} - \boldsymbol{a}),$$

resulting in the intensity function

$$I_{A_q}(\boldsymbol{\xi}) = \frac{1}{q^{2n}} \left| \sum_{\boldsymbol{a} \in A_q} e^{2\pi i \boldsymbol{a} \cdot \boldsymbol{\xi}} \right|^2 .$$

It is further more convenient however (due to the fact that the diameter of A_q is $O(q)$ rather than 1) to multiply the intensity by q^n, so we shall further refer to its definition as follows

$$I_{A_q}(\boldsymbol{\xi}) = \frac{1}{q^n} \left| \sum_{\boldsymbol{a} \in A_q} e^{2\pi i \boldsymbol{a} \cdot \boldsymbol{\xi}} \right|^2 = \frac{1}{q^n} \sum_{\boldsymbol{a}, \boldsymbol{a}' \in A_q} e^{2\pi i (\boldsymbol{a} - \boldsymbol{a}') \cdot \boldsymbol{\xi}}. \tag{2.1}$$

2.1 Averaging

We are now ready to introduce different types of averages of the intensity function. First consider the so-called *solid average*: for $R \gg 1$ let

$$\bar{I}_{A_q} = \frac{1}{c_n R^n} \int_{R \le \|\boldsymbol{\xi}\| \le 2R} I_{A_q}(\boldsymbol{\xi}) d\boldsymbol{\xi} \approx \frac{1}{R^n} \int I_{A_q}(\boldsymbol{\xi}) \psi(\boldsymbol{\xi}/q) d\boldsymbol{\xi}. \tag{2.2}$$

The constant c_n above is the volume of the unit ball in $\mathbb{R}^n$; in the sequel the value of c_n may vary, but will depend on the dimension only. For simplicity we shall suppress such constants (not characteristic of the sample set E; in particular this includes constants appearing after integration) by using the $\approx$ symbol instead of the equality (e.g. $\int_0^R r^{n-1} dr \approx R^n$). In the same fashion we shall use the symbols $\lesssim$ and $\gtrsim$ to suppress such constants in inequalities. In addition, the integration in (2.2) has been extended to the whole $\mathbb{R}^n$ by incorporating a radially symmetric (henceforth we will simply say *radial*) infinitely differentiable function[2], which is identically equal to 1 in the spherical shell $\|\boldsymbol{x}\| \in [1,2]$ and vanishes outside a slightly larger shell, say $\|\boldsymbol{x}\| \in [.9, 1.1]$. Writing out the definition of I_{A_q} in (2.2), we see that

$$\bar{I}_{A_q} = q^{-n} \sum_{\boldsymbol{a}, \boldsymbol{a}' \in A_q} \widehat{\psi}(R(\boldsymbol{a} - \boldsymbol{a}')), \tag{2.3}$$

[2]Cutoff C^∞ functions are routinely used further in the paper, always being denoted as ψ. One can also use Gaussians as cutoff functions. In both cases the "tails" beyond the effective supports of the functions themselves or their Fourier transforms are negligible and will not be estimated.

where

$$\widehat{\psi}(\boldsymbol{\xi}) = \int_{\mathbb{R}^n} e^{-2\pi i \boldsymbol{x}\cdot\boldsymbol{\xi}} \psi(\boldsymbol{x}) d\boldsymbol{x},$$

the Fourier transform of ψ. By the uncertainty principle, $\widehat{\psi}(R(\boldsymbol{a}-\boldsymbol{a}'))$ is concentrated in the set

$$\{(\boldsymbol{a}, \boldsymbol{a}') \in A_q \times A_q : \|\boldsymbol{a}-\boldsymbol{a}'\| \leq R^{-1}\}.$$

That, for large R, leaves out the case $\boldsymbol{a} \neq \boldsymbol{a}'$ in the double sum in (2.3).

We see from the expression (2.3) that

$$\bar{I}_{A_q} \approx q^{-n} \cdot \# A_q \approx 1,$$

As far as the solid average is concerned, all Delaunay sets of the same density are the same.

In order to obtain a more interesting dependence on geometry, consider the *spherical average*

$$\sigma_{A_q}(r) = \int I_{A_q}(r\boldsymbol{x}) d\omega_{\boldsymbol{x}}, \tag{2.4}$$

where the integral is taken with respect to the surface measure on the unit sphere, cf. (1.3).

We now average over the length of the wave vector over the relevant range $\|\boldsymbol{\xi}\| \leq q$. By the uncertainty principle, this would correspond to the case when the actual atom size is of order q^{-1} (on the scale when the atomic spacing is of order 1). As we have mentioned earlier, the quantity $\sigma_{A_q}(r)$ is not interesting for small r, so we can once again fix a smooth cut-off function ψ (this time of one variable on the non-negative real axis), such that $\psi(r) \equiv 1$ for $r \in [0, 1]$ and vanishes for $r > 1.1$. Let the *second moment*

$$M_{A_q,2} = \frac{1}{q^n} \int_0^\infty \sigma^2_{A_q}(r) \widehat{\psi}(r/q) r^{n-1} dr, \tag{2.5}$$

cf. (1.4) Observe that the presence of the cutoff function ψ in the integral effectively means that integration is performed up to $r \approx q$, hence the factor $\frac{1}{q^n}$ plays the normalization role.

In order to understand the meaning of the second moment, let us introduce the *distance multiplicity function* by the formula

$$m_{\frac{1}{q}}(r) = \# \left\{ (\boldsymbol{a}, \boldsymbol{a}') : r \leq \|\boldsymbol{a}-\boldsymbol{a}'\| \leq r + \frac{1}{q} \right\}. \tag{2.6}$$

The main result in this section is the following theorem.

Theorem 2.2. *We have*

$$\begin{aligned} M_{A_q,2} &\approx 1 + G_q, \quad \textit{where} \\ G_q &= \tfrac{1}{q^{3n-1}} \textstyle\sum_r \left(\frac{m_{\frac{1}{q}}(r)}{r^{\frac{n-1}{2}}} \right)^2, \end{aligned} \tag{2.7}$$

with the sum being taken over a maximal $\frac{1}{q}$-separated subset of $\Delta(A_q) \backslash 0$.

Before we present the proof of Theorem 2.2, let us point out that the quantity under the summation sign is calculated as follows. For each $\boldsymbol{a} \in A_q$ one takes a spherical shell of radius r and thickness $\frac{1}{q}$ centered at the point $\boldsymbol{a}$ and counts the number of elements of A_q in this shell, weighted by $\frac{1}{r^{\frac{n-1}{2}}}$. Then the summation over the choice of the center $\boldsymbol{a}$ is effected. Further, the result gets squared and summed over all r's which are multiples of $\frac{1}{q}$.

The formula (2.7) shows that $M_{A_q,2}$ tends to get larger when the distance multiplicity function $m_{\frac{1}{q}}(r)$ deviates a lot from its expected value, which as it's easy to see is approximately $q^n \cdot (q^{-1} r^{n-1})$. Hence, if the set A_q is random, one can expect that

$$G_q \approx q^{-3n+1} \cdot q^{2n-2} \cdot q \int_0^q r^{n-1} dr \approx 1. \tag{2.8}$$

However, if A has a translation-invariant structure, the distance multiplicity function $m_{\frac{1}{q}}(r)$ can deviate from its expected value considerably, at least for $n = 2, 3$.

Remark 2.3. In particular, if $A = \mathbb{Z}^n$, it is well known that the quantity $q^{-n} m_{\frac{1}{q}}(r)$ can be as large as some quantity $d_n(r)$, which for $n = 2$ equals the maximum number of divisors an integer r can have (which is asymptotically smaller any positive power of r but greater than any power of $\log r$). For $n = 3$ it cam be as large as $r \log r$. For $n = 4$ it is $O(r^2)$. For $n \geq 5$ it is known that if a sphere of radius r has an integer point on it, the total number of points of $\mathbb{Z}^n$ is guaranteed to be $\approx r^{n-2}$. See [11] and [7] for details.

For the practical purpose of processing the powder diffraction data, an experimental outcome when the quantity (2.7) is fairly small, should signal the lack of translation-invariant structure in the sample. On the other hand, different types of crystal structure should yield different values of the second moment. In particular, its value for the cubical lattice largely exceeds the value one gets for the tetrahedral lattice. The latter, although optimal from the packing point of view is far from optimal from the point of view of the distance multiplicity function second moment.

2.2 Proof of Theorem 2.2

We have

$$\begin{aligned} \sigma_{A_q}(r) &= \int I_{A_q}(r\boldsymbol{x}) d\omega_{\boldsymbol{x}} \\ &= c_n + q^{-n} \sum_{\boldsymbol{a} \neq \boldsymbol{a}'} \int e^{2\pi i (\boldsymbol{a}-\boldsymbol{a}') \cdot r\boldsymbol{x}} d\omega_{\boldsymbol{x}} \\ &= c_n + q^{-n} \sum_{\boldsymbol{a} \neq \boldsymbol{a}'} \widehat{\omega}(r(\boldsymbol{a}-\boldsymbol{a}')). \end{aligned} \tag{2.9}$$

Henceforth, $\widehat{\omega}$ denotes the Fourier transform of the surface measure on the unit sphere.[3] Note that the first term corresponds to the choice $\boldsymbol{a} = \boldsymbol{a}'$ in the intensity definition (2.1). The constant c_n is now the surface area of the unit sphere, to be absorbed into the $\approx$ symbol, i.e we'll further assume $\widehat{\omega}(\mathbf{0}) = 1$.

It follows that

$$\begin{aligned} M_{A_q,2} &\approx 1 + q^{-2n} \sum_{\boldsymbol{a}\neq\boldsymbol{a}'} \int_0^\infty \widehat{\omega}(r(\boldsymbol{a}-\boldsymbol{a}'))r^{n-1}\psi(r/q)dr \\ &+ q^{-3n} \sum_{\boldsymbol{a}\neq\boldsymbol{a}';\boldsymbol{b}\neq\boldsymbol{b}'} \int_0^\infty \widehat{\omega}(r(\boldsymbol{a}-\boldsymbol{a}'))\widehat{\omega}(r(\boldsymbol{b}-\boldsymbol{b}'))\psi^2(r/q)r^{n-1}dr \\ &= 1 + Y_q + X_q. \end{aligned}$$

To evaluate that quantity Y_q let us view the test function ψ as a radial function of a variable $\boldsymbol{\xi} \in \mathbb{R}^n$, rewrite the integral for Y_q as an integral over $\mathbb{R}^n$ and apply Plancherel's theorem

$$\int \widehat{f}(\boldsymbol{\xi})\widehat{g}(\boldsymbol{\xi})d\boldsymbol{\xi} = \int f(\boldsymbol{x})g(\boldsymbol{x})d\boldsymbol{x}.$$

and the convolution theorem

$$\widehat{f}\widehat{g} = \widehat{f*g}, \quad \text{where} \quad f*g(\boldsymbol{x}) = \int f(\boldsymbol{x}-\boldsymbol{y})g(\boldsymbol{y})d\boldsymbol{y}.$$

We get

$$\begin{aligned} q^{2n}Y_q &\approx \sum_{\boldsymbol{a}\neq\boldsymbol{a}'} \int \widehat{\omega}(\boldsymbol{\xi}\|\boldsymbol{a}-\boldsymbol{a}'\|)\psi(\boldsymbol{\xi}/q)d\boldsymbol{\xi} \\ &= \sum_{\boldsymbol{a}\neq\boldsymbol{a}'} \frac{q^n}{\|\boldsymbol{a}-\boldsymbol{a}'\|^{n-1}} \int\int \left(\int \widehat{\psi}[q(\boldsymbol{x}-\boldsymbol{y})]d\omega_{\|\boldsymbol{a}-\boldsymbol{a}'\|^{-1}\boldsymbol{y}}\right)d\boldsymbol{x} \end{aligned} \tag{2.10}$$

Above, $d\omega_{\|\boldsymbol{a}-\boldsymbol{a}'\|^{-1}\boldsymbol{y}}$ is the surface measure on the sphere of radius $\|\boldsymbol{a}-\boldsymbol{a}'\|$. Hence, we integrate first in $\boldsymbol{x}$, and for each $\boldsymbol{y}$ we have the following situation. By the uncertainty principle, the Fourier transform of the test function $\psi(\boldsymbol{\xi})$ is a radial function which vanishes faster than any inverse power of the distance from the origin. Effectively, one can substitute $\widehat{\psi}$ by the characteristic function of the unit ball. Then, for each $\boldsymbol{y}$, integration in $\boldsymbol{x}$ can be effectively restricted to the ball of radius $\frac{1}{q}$ around $\boldsymbol{y}$. Therefore,

$$\begin{aligned} q^{2n}Y_q &\approx \sum_{\boldsymbol{a}\neq\boldsymbol{a}'} \frac{1}{\|\boldsymbol{a}-\boldsymbol{a}'\|^{n-1}} \int d\omega_{\|\boldsymbol{a}-\boldsymbol{a}'\|^{-1}\boldsymbol{y}} \\ &= \sum_{\boldsymbol{a}\neq\boldsymbol{a}'} 1 \approx q^{2n}. \end{aligned} \tag{2.11}$$

[3] A calculation yields

$$\widehat{\omega}(\boldsymbol{\xi}) = 2\pi \frac{J_{\frac{n}{2}-1}(\|\boldsymbol{\xi}\|)}{\|\boldsymbol{\xi}\|^{\frac{n-1}{2}}},$$

where $J_{\frac{n}{2}-1}$ is the Bessel function of order $\frac{n}{2}-1$. However, this formula will not be used.

It follows that $Y_q = O(1)$.

The quantity X_q is evaluated in the same way. To simplify matters, there is no harm writing $\psi(r/q)$ instead of $\psi^2(r/q)$ in the integral for X_q, by the properties of the test function ψ. Then writing the integral as an integral over $\mathbb{R}^n$ and applying the Plancherel theorem and the convolution theorem we have

$$\begin{aligned} q^{3n}X_q &= \sum_{\boldsymbol{a}\neq\boldsymbol{a}',\,\boldsymbol{b}\neq\boldsymbol{b}'} \int \left(\widehat{\omega}[\|\boldsymbol{a}-\boldsymbol{a}'\|\boldsymbol{\xi}]\psi(\boldsymbol{\xi}/q)\right)^{\wedge}(\boldsymbol{x}) \cdot \left(\widehat{\omega}[\|\boldsymbol{b}-\boldsymbol{b}'\|\boldsymbol{\xi}]\right)^{\wedge}(\boldsymbol{x})d\boldsymbol{x} \\ &= \sum_{\boldsymbol{a}\neq\boldsymbol{a}',\,\boldsymbol{b}\neq\boldsymbol{b}'} \frac{1}{(\|\boldsymbol{a}-\boldsymbol{a}'\|\|\boldsymbol{b}-\boldsymbol{b}'\|)^{n-1}} \int [d\omega_{\|\boldsymbol{a}-\boldsymbol{a}'\|^{-1}\boldsymbol{y}} * (q^n\psi(q\boldsymbol{y}))](\boldsymbol{x}) d\omega_{\|\boldsymbol{b}-\boldsymbol{b}'\|^{-1}\boldsymbol{x}} \qquad (2.12) \\ &\approx \sum_{\boldsymbol{a}\neq\boldsymbol{a}',\,\boldsymbol{b}\neq\boldsymbol{b}'} \frac{1}{(\|\boldsymbol{a}-\boldsymbol{a}'\|\|\boldsymbol{b}-\boldsymbol{b}'\|)^{n-1}} \int_{\mathcal{S}(\|\boldsymbol{a}-\boldsymbol{a}'\|,\frac{1}{q})} q\, d\omega_{\|\boldsymbol{b}-\boldsymbol{b}'\|^{-1}\boldsymbol{x}}. \end{aligned}$$

Above, $\mathcal{S}(\|\boldsymbol{a}-\boldsymbol{a}'\|,\frac{1}{q})$ denotes a spherical shell of radius $\|\boldsymbol{a}-\boldsymbol{a}'\|$ and thickness $\frac{1}{q}$.

Indeed, $\omega_{\|\boldsymbol{a}-\boldsymbol{a}'\|^{-1}\boldsymbol{y}}$ is the surface measure on the sphere of radius $\|\boldsymbol{a}-\boldsymbol{a}'\|$, and its convolution with $q^n\psi(q\boldsymbol{y})$ (which as in has been done in the estimate for Y_q, can be thought to be supported in the ball of radius $\frac{1}{q}$ and apart from that integrates into approximately one) is effectively a density, supported in the shell $\mathcal{S}(\|\boldsymbol{a}-\boldsymbol{a}'\|,\frac{1}{q})$, of the value of approximately q. As $d\omega_{\|\boldsymbol{b}-\boldsymbol{b}'\|^{-1}\boldsymbol{x}}$ is the surface measure on the sphere of radius $\|\boldsymbol{b}-\boldsymbol{b}'\|$, the integral in the last line of (2.12) will be non-zero only if

$$\big|\|\boldsymbol{a}-\boldsymbol{a}'\| - \|\boldsymbol{b}-\boldsymbol{b}'\|\big| \lesssim \frac{1}{q}.$$

If this is the case, the integral will be approximately $q\|\boldsymbol{a}-\boldsymbol{a}'\|^{n-1}$, and we can set $\|\boldsymbol{a}-\boldsymbol{a}'\| = \|\boldsymbol{b}-\boldsymbol{b}'\| = r$ in the pre-integration factor, as far as the formula (2.7) is concerned. Furthermore, for each r, which can be assumed to run a discrete sequence of values, which are multiples of $\frac{1}{q}$, up to $r = q$, is nothing but the square of the quantity $m_{\frac{1}{q}}(r)$, which has been defined by (2.6). This proves Theorem 2.2.$\Box$

2.3 Connections with geometric combinatorics

We start out by briefly recalling basic definitions from the theory of distance sets. See, for example, [14] and the references contained therein for a thorough treatment of this old and beautiful subject.

Let A be a Delaunay set. Define $\Delta(A_q)$, following (1.5). The *Erdös Distance Conjecture* (EDC) says that

$$\#\Delta(A_q) \gtrapprox q^2, \qquad (2.13)$$

where # gives cardinality of a finite set. Here and throughout the paper, $X \lessapprox Y$ with respect to the controlling parameter q means that for every $\epsilon > 0$ there exists a constant $C_\epsilon > 0$ such that $X \le C_\epsilon q^\epsilon Y$. Accordingly, $X \gtrapprox Y$ means that for every $\epsilon > 0$ there exists a constant $c_\epsilon > 0$

such that $X \geq c_\epsilon q^{-\epsilon} Y$. In the case when $A = \mathbb{Z}^n$, the classic number theory calculation (see for example [11] and [7]) shows that

$$\#\Delta(A_q) \approx \frac{q^2}{\sqrt{\log(q)}} \text{ when } n = 2,$$

and

$$\#\Delta(A_q) \approx q^2 \text{ when } n \geq 3.$$

In particular, in the case $n = 2$ the squares of the distances are those integers in the interval $[0, 2q^2]$, which can be represented as sums of two squares. The average density of such integers in the interval above is proportional to $\frac{q^2}{\sqrt{\log(q)}}$.

A possible approach to EDC, exploited by several authors in recent years, is the following. Let

$$m(r) = \#\{(\boldsymbol{a}, \boldsymbol{a'}) \in A_q \times A_q : \|\boldsymbol{a} - \boldsymbol{a'}\| = r\}, \tag{2.14}$$

cf. (2.6).

Suppose that we could show that

$$\sum_{r \in \Delta(A_q) \setminus 0} m^2(r) r^{-(n-1)} \lessapprox q^{3n-1}. \tag{2.15}$$

Since

$$\sum_{r \in \Delta(A_q) \setminus 0} m(r) \approx q^{2n}, \tag{2.16}$$

by definition of $m(r)$, Cauchy-Schwartz inequality implies that

$$\begin{aligned} q^{4n} &\approx \left(\textstyle\sum_{r \in \Delta(A_q) \setminus 0} m(r) \cdot 1\right)^2 \\ &\leq \textstyle\sum_{r \in \Delta(A_q) \setminus 0} 1 \cdot \sum_{r \in \Delta(A_q) \setminus 0} m^2(r) [q^{n-1} r^{-(n-1)}] \\ &\lessapprox q^{4n-2} \cdot \#\Delta(A_q), \end{aligned} \tag{2.17}$$

and EDC follows.

Equivalently, in the notation of Theorem 2.2, if we assume that $M_{A_q,2} \lessapprox 1$, we are guaranteed to have $\gtrapprox q^2$ distinct $\frac{1}{q}$-separated distances in the distance set $\Delta(A_q)$.

2.4 Maximum Intensity Conjecture

Theorem 2.2 and the argument in the estimates (2.15–2.17) in the previous section suggest that in the accuracy level built into the $\lessapprox$, $\gtrapprox$ symbols (i.e. more accurately than up to an arbitrarily small positive power of the controlling parameter q) the distance set estimate (2.13) is equivalent to the appropriate upper bound

$$M_{A_q,2} \lessapprox 1. \tag{2.18}$$

The quantity $M_{A_q,2}$ can be computed explicitly in cases when the underlying set A possesses translational symmetry. Further in this section we shall do it for the lattice $\mathbb{Z}^n$.

This leads us to conjecture that in the the class $\mathcal{A}$ of Delaunay sets under consideration, one should have $M_{A_q,2} \lessapprox 1$, and

$$\sup_{A\in\mathcal{A}} M_{A_q,2} = M_{\mathbb{Z}_q^n,2}, \tag{2.19}$$

where $\mathbb{Z}_q^n = \mathbb{Z}^n \cap [0,q]^n$.

We shall further refer to this conjecture as the *Maximum Intensity Conjecture* (MIC). It says that if we bombard a crystalline powder with X-rays, the second moment defined by (1.4) of the powder diffraction intensity will be the greatest if the identical atoms of the crystal were arranged in the nodes of the cubic lattice. The Maximum Intensity Conjecture would immediately lead to the resolution of the Erdös Distance Conjecture for Delaunay sets, and many other problem in geometric combinatorics and analysis.

If we accept the Maximum Intensity Conjecture, we can deduce the EDC if we can show that

$$M_{\mathbb{Z}_q^n,2} \lessapprox 1. \tag{2.20}$$

This fact results immediately from Theorem 2.2, the calculation (2.8) that follows it and Remark 2.3. Note that according to Remark 2.3, in dimensions 4 and higher one would have a $\lesssim$ estimate instead of $\lessapprox$. It is not clear whether for $n=3$ one can strengthen (2.20) to $M_{\mathbb{Z}_q^n,2} \lesssim 1$. On the one hand, the best one can prove is that $M_{\mathbb{Z}_q^3,2} \lesssim \log q$ (this follows from Remark 2.3). On the other hand, it is known that $\#\Delta(\mathbb{Z}_q^3) \gtrsim q^2$. Hence it is not clear whether $\log q$ is intrinsic or is an artifact of the Cauchy-Schwartz inequality application in (2.17) and the supremum estimate $q^3 r \log r$ for the quantity $m_{\frac{1}{q}}(r)$.

As a matter of fact, we can prove a stronger statement:

$$\begin{aligned} \sigma_{A_q}(r) &\lesssim \tfrac{q}{r}, \quad \text{for } r \geq 1,\ n \geq 4, \\ \sigma_{A_q}(r) &\lessapprox \tfrac{q}{r}, \quad \text{for } r \geq 1,\ n = 2,3. \end{aligned} \tag{2.21}$$

First off, note that from (2.9), for $r \lesssim \frac{1}{q}$ we have $\sigma_{A_q}(r) \approx q^n$. Observe that the contribution of the interval $r \lesssim \frac{1}{q}$ into (2.5) is then $O(1)$.

Now let us assume $r \gtrsim \frac{1}{q}$. Return to (2.9) where we can drop the first term and use the translational invariance of the lattice to reduce the double sum to a one index sum as follows:

$$\sigma_{\mathbb{Z}_q^n}(r) \approx \sum_{\boldsymbol{a}\in\mathbb{Z}_q^n} \widehat{\omega}(r\boldsymbol{a}) \approx \sum_{\boldsymbol{a}\in\mathbb{Z}^n} \widehat{\omega}(r\boldsymbol{a})\widehat{\psi}(\boldsymbol{a}/q). \tag{2.22}$$

Above, we have extended the summation over the whole lattice $\mathbb{Z}^d$ by using a radial test function ψ, which is supported in the unit ball and is identically one in unit ball of radius $\frac{1}{3}$. Without loss of generality $\widehat{\psi}(\boldsymbol{\xi})$ can be though non-negative (e.g. ψ can be chosen as a convolution of some radial function with itself) and by the uncertainty principle it vanishes faster than any power of $\|\boldsymbol{\xi}\|$ beyond the unit ball. Now we can apply the Poisson summation formula[4]

$$\sum_{\boldsymbol{a}\in\Lambda}\psi(\boldsymbol{a})=\frac{1}{|\Lambda|}\sum_{\boldsymbol{a}\in\Lambda^*}\widehat{\psi}(\boldsymbol{a}),$$

where Λ is any lattice, Λ^* is the reciprocal lattice and $|\Lambda|$ is the volume of the basic element of the lattice. Using the convolution theorem we obtain

$$\sigma_{\mathbb{Z}_q^n}(r)\lesssim\frac{1}{r^{n-1}}\sum_{\boldsymbol{a}\in\mathbb{Z}^n}\int q^d\psi[q(\boldsymbol{a}-\boldsymbol{x})]d\omega_{r^{-1}\boldsymbol{x}}\approx\frac{q}{r^{n-1}}\#\{\boldsymbol{a}\in\mathbb{Z}^n:\,\|\boldsymbol{a}\|=r\}. \tag{2.23}$$

Remark 2.3 gives one the upper bound for the number of the lattice points on the sphere of radius r. Namely

$$\#\{\boldsymbol{a}\in\mathbb{Z}^n:\,\|\boldsymbol{a}\|=r\}\ \lessapprox\ r^{n-2},$$

and in $n\geq 4$ one can strengthen the estimate with $\lesssim$ rather than $\lessapprox$. This proves the estimate (2.21) which in turn immediately implies that

$$\begin{aligned} M_{A_q,2}&\ \lesssim\ 1,\quad\text{for}\ \ n\geq 4,\\ M_{A_q,2}&\ \lessapprox\ 1,\quad\text{for}\ \ n=2,3. \end{aligned} \tag{2.24}$$

2.5 Anysotropic distances

We would like to briefly point out an issue that instead of averaging with respect to the unit sphere S^{n-1} in (1.3) and (2.4) one might consider anisotropic averages, defined as follows.

Let $K\in\mathbb{R}^n$ be a strictly convex body of unit volume, with the smooth boundary ∂K and such that the curvature on ∂K is bounded away from zero. The in (1.3) and (2.4) above we can think that $d\omega_{\boldsymbol{x}}$ is the surface measure on ∂K, thus speaking about K-averages and moments (1.4, 2.5). Note that the reciprocal body K^* to K is defined as

$$\{\boldsymbol{\xi}\in\mathbb{R}^n:\,\sup_{\boldsymbol{x}\in K}\boldsymbol{\xi}\cdot\boldsymbol{x}\leq 1\}.$$

(I.e in the direction $\boldsymbol{d}$, the boundary ∂K is $\frac{1}{\|\boldsymbol{x_d}\|}$ away from the origin, where $\boldsymbol{x_d}$ is the point on ∂K where the direction of the normal is $\boldsymbol{d}$.)

[4]Have we been dealing with some other lattice other than $\mathbb{Z}^n$ (which is self-dual) the summation in (2.23) would be taken over the reciprocal lattice

We would like to point out that the analytical machinery of this and the subsequent section does apply to this situation with the following changes.

i. The constants absorbed in the $\approx$, etc. symbols now depend on K.

ii. Theorem 2.2 holds, except in the multiplicity function (2.6, 2.14) one should change $\|\boldsymbol{a}-\boldsymbol{a}'\|$ to $\|\boldsymbol{a}-\boldsymbol{a}'\|_{K^*}$, where $\|\mathbf{0}\|_K = 0$ and for $\boldsymbol{x} \neq \mathbf{0}$,

$$\|\boldsymbol{x}\|_K = r : \, r^{-1}\boldsymbol{x} \in \partial K.$$

iii. Despite Remark 2.3 no longer applies and the point-wise estimate (2.21) can no longer be established (one can construct explicit counterexamples at least in $n = 2$), the conclusion (2.24) of the preceding section still holds, although it requires a more intricate proof, which goes through if $n \geq 3$, without using number theory. The dimension $n = 2$, for a general K, is an open problem however.

For details, see [9], [10] and the references contained therein. Note in particular that the case of the class of convex bodies K used in place of the unit sphere automatically covers the situation when $K = S^{n-1}$, but any other parallelepipedal lattice is used in Section 2.4 instead of $\mathbb{Z}^n$.

3 Continuous case

Let us now discuss the continuous situation. The general set-up is the compact set $E \subset [0,1]^n$, of dimension $\alpha \in (0, n)$. To give a "physical" definition of dimension, suppose that there exists a measure μ supported on E, such that for any $\boldsymbol{x}$ in the support of μ, one has

$$\mu(B(\boldsymbol{x}, \delta)) \approx \delta^\alpha, \tag{3.1}$$

for all sufficiently small $\delta > 0$; the notation $B(\boldsymbol{x}, \delta)$ stands for a n-dimensional ball of the radius δ centered at $\boldsymbol{x}$.

Physically however, $\delta \to 0$ should mean that $\delta \geq \delta_0$, the latter being, say the atomic size. From this point of view, the set $E = \frac{1}{q}A_q$ (i.e A_q, which gets scaled into the unit cube) discussed in the previous section has dimension $\alpha = \frac{n}{2}$. Indeed, we can construct the measure μ as follows. Let $\psi(\boldsymbol{x})$ be the characteristic function of the unit ball. Let

$$d\mu_q(\boldsymbol{x}) = \frac{1}{q^n} \sum_{\boldsymbol{a} \in \frac{1}{q}A_q} [q^{2n}\psi(q^2\boldsymbol{x})] d\boldsymbol{x}. \tag{3.2}$$

Then $\int d\mu_q(\boldsymbol{x}) \approx 1$ and it is an easy calculation to verify that the definition (3.1) is satisfied with $\alpha = \frac{n}{2}$, for $\delta \gtrsim \frac{1}{q^2}$. Recall that this dimension corresponded to "thickening" each point of A_q into a ball of radius $\approx \frac{1}{q}$, or the "physical" assumptions that atomic sizes are some q

times smaller that the spacing between the atoms. If one substitutes $[q^{2n}\psi(q^2\boldsymbol{x})]$ in the formula (3.2) with $q^{\frac{n^2}{\alpha}}\psi(q^{\frac{n}{\alpha}}\boldsymbol{x})$, one would get a "fractal" measure of dimension α in the sense that the definition (3.1) holds for all $\delta \gtrsim q^{-\frac{n}{\alpha}}$. Moreover, if one denotes E_q the support of the measure μ_q, then after taking a sequence of $q's$, which rapidly enough goes to infinity, the set $\bigcap_q E_q$ will represent a bona fide fractal of dimension α, with the bona fide fractal measure μ_∞ on it (obtained as a limit of the measures μ_q), which satisfies (3.1) for all $1 > \delta > 0$.

Conversely, if one takes the fractal measure μ_∞ and convolves it with the quantity $q^{\frac{n^2}{\alpha}}\psi(q^{\frac{n}{\alpha}}\boldsymbol{x})$ (which integrates approximately into one) the density resulting from the convolution will satisfy (3.1), but only for $\delta \gtrsim q^{-\frac{n}{\alpha}}$. See [4], [8] for details.

3.1 Falconer distance problem

Returning to the general fractal dimensional set up, from (3.1) it follows that the α-*energy*

$$I_\alpha(\mu) = \int\int \frac{d\mu_{\boldsymbol{x}} d\mu_{\boldsymbol{y}}}{\|\boldsymbol{x}-\boldsymbol{y}\|^\alpha} = O(1). \tag{3.3}$$

Rewriting (3.3) by Plancherel's theorem and passing to polar coordinates we obtain

$$I_\alpha(\mu) = \int \frac{|\widehat{\mu}(\boldsymbol{\xi})|^2}{\|\boldsymbol{\xi}\|^{n-\alpha}} = \int_0^\infty r^{\alpha-1}\sigma_E(r)dr, \tag{3.4}$$

where $\sigma_E(r)$ has been defined by (1.3) and models the observable intensity in power diffraction experiments.

It follows that on average $\sigma_E(r)$ is smaller than $r^{-\alpha}$. However, it does not necessarily happen for all r, as the examples constructed by Sjölin ([18] and the authors ([10]) indicate.

By compactness of $E \subset [0,1]^d$ it follows that the expression (3.3) will not change if we multiply, say $d\mu_{\boldsymbol{x}}$ in (3.3) by a cutoff function $\psi(\boldsymbol{x})$ which equals 1 on $[0,1]^d$ and is supported on a slightly larger set. By the convolution theorem and the uncertainty principle, this implies that for $r \gg 1$

$$\sigma_E(r) \approx \frac{1}{r^{n-1}} \int r^{n-\alpha}\frac{|\widehat{\mu}(\boldsymbol{\xi})|^2}{\|\boldsymbol{\xi}\|^{n-\alpha}}\chi_{\mathcal{S}(r,1)}(\boldsymbol{\xi})d\boldsymbol{\xi} \lesssim r^{1-\alpha}I_\alpha(\mu).$$

Above, $\chi_{\mathcal{S}(r,1)}$ denotes the characteristic function of the spherical shell $\mathcal{S}(r,1)$ of radius r and thickness 1.

Hence, one has the general bound

$$\sigma_E(r) \lesssim r^{-\alpha+1}. \tag{3.5}$$

It turns out that the quantity $\sigma_E(r)$ is closely related to the distance set $\Delta(E)$. To this effect, there is a continuous analog of the Erdös distance conjecture, known as the *Falconer distance* *problem*, which was first formulated in ([5]). It states that that if $\alpha > \frac{n}{2}$, then the Lebesgue measure of the distance set $\Delta(E)$ is positive.

The relation between the spherical average $\sigma_E(r)$ and the distance set $\Delta(E)$ gets established via the machinery developed by Mattila ([12]). The discussion in the previous section presented in fact a discrete analog of this machinery; the continuous situation can be summarized as follows.

Mattila proves that for $\alpha \geq \frac{n}{2}$, if

$$M_{E,2} = \int_0^\infty \sigma_E^2(r) r^{n-1} dr < \infty, \tag{3.6}$$

then the Lebesgue measure of $\Delta(E)$ is positive, using the continuous version of the argument in (2.14-2.17).

Mattila considers the pull-forward ν on $\Delta(E)$ of the measure $\mu \times \mu$ on $E \times E$, under the distance map, defined as follows: for a test function $\psi(r)$ on the non-negative real axis,

$$\int \psi(r) d\nu_r = \int \int \psi(\|\boldsymbol{x} - \boldsymbol{y}\|) d\mu_{\boldsymbol{x}} d\mu_{\boldsymbol{y}}.$$

Then by the Cauchy-Schwartz inequality and Plancherel theorem,

$$1 \lesssim \left(\int d\nu_r \right)^2 \leq |\Delta(E)| \cdot \int |\widehat{\nu}(t)|^2 dt, \tag{3.7}$$

as long as ν has an L^2 density. $|\Delta(E)|$ above denotes the Lebesgue measure of the distance set.

Mattila then shows that if $\widehat{\nu}$ denotes the one-dimensional Fourier transform of the distance set measure ν, it turns out that

$$\widehat{\nu}(t) \approx t^{\frac{n-1}{2}} \int |\widehat{\mu}(\boldsymbol{\xi})|^2 d\omega_{\boldsymbol{\xi}}. \tag{3.8}$$

I.e. *the spherical average represents a weighted Fourier transform of the distance measure,* whose discrete analog is the distance multiplicity function $m(r)$, cf. (2.14). See [12] for details.

Observe that rewriting the integral (3.6) as an integral over $\mathbb{R}^n$ and using the bound (3.5) for $\sigma_E(r)$ it follows that

$$\begin{aligned} M_{E,2} &\lesssim \int |\widehat{\mu}(\boldsymbol{\xi})|^2 \|\boldsymbol{\xi}\|^{-n+(n-\alpha+1)} d\boldsymbol{\xi} \\ &\approx \int\int \|\boldsymbol{x} - \boldsymbol{y}\|^{-(n-\alpha+1)} d\mu_{\boldsymbol{x}} d\mu_{\boldsymbol{y}} \\ &\equiv I_{n+1-\alpha}(\mu). \end{aligned} \tag{3.9}$$

This answers affirmatively to the question posed in the Falconer distance problem, provided that $\alpha \geq \frac{n+1}{2}$.

The question arises whether the improvement towards the desired $\alpha > \frac{n}{2}$ can be gained by strengthening the bound (3.5).

The best known result in this direction is due to Wolff ([19]), in $n = 2$, and Erdoğan ([2]) in higher dimensions. The result implies that the Lebesgue measure of $\Delta(E)$ is indeed positive if the Hausdorff dimension of E is greater than $\frac{n}{2} + \frac{1}{3}$.

It is based on the following estimate:

$$\sigma_E(r) \lesssim r^{-\beta}, \ \ \forall \beta \ < \begin{cases} \alpha, & \text{for} \quad \alpha \leq \frac{n-1}{2}, \\ \frac{n-1}{2} & \text{for} \ \ \frac{n-1}{2} \leq \alpha \leq \frac{n}{2}, \\ \frac{n+2\alpha-2}{4} & \text{for} \ \ \frac{n}{2} \leq \alpha \leq \frac{n+1}{2}. \end{cases} \tag{3.10}$$

It is interesting that due to a counterexample of Sjölin ([18]) the bounds (3.10) cannot be improved in $n = 2$. However, they are are not likely to be optimal for $\alpha > \frac{n-1}{2}$ and $n \geq 3$. For $n = 3$ the same counterexample shows that the Falconer conjecture cannot be resolved by improving the bound (3.10) alone. However, it may be possible (see [2]) in dimensions $n \geq 4$, cf. Remark (2.3).

3.2 On additive structure of measures

We have shown earlier that the second moment $M_{E,2}$ is indicative of the distance structure of the set E. In this final section we would like to discuss how higher moments are indicative of the presence of additive, or translation-invariant structure and translational symmetry and therefore should represent a certain signature thereof.

We shall content ourselves with the fourth moment $M_{E,4}$ only and point out two principal facts:

i. Large $M_{E,4}$ indicate the presence of an additive structure;

ii. The total lack of additive structure results in very small values of $M_{E,4}$; in particular the Falconer conjecture holds for sets with the total lack of additive structure.

Let us first show how the fourth moment is indicative of translations. Let us confine ourselves to the critical case $\alpha = \frac{n}{2}$.

Define $\boldsymbol{a} \approx_\delta \boldsymbol{b}$ if $\|\boldsymbol{a} - \boldsymbol{b}\| \leq \delta$. Let $X = (\boldsymbol{x}_1, \boldsymbol{x}_2) \in \mathbb{R}^{2n}$, $Y = (\boldsymbol{y}_1, \boldsymbol{y}_2) \in \mathbb{R}^{2n}$ and $\mu^* = \mu_X \times \mu_Y = \mu \times \mu \times \mu \times \mu$.

Suppose, ψ is a radial cut-off function which is supported in the spherical shell $\{\boldsymbol{\xi} : .9 \leq \|\boldsymbol{\xi}\| \leq 2.1\}$, and is identically one for $1 \leq \|\boldsymbol{\xi}\| \leq 2$. Let $R \gg 1$. Consider the following variation of the fourth moment:

$$M_{E,4}(R) = \int_{R \leq r \leq 2R} \sigma_E^4(r) r^{n-1} dr = \int_{R \leq \|\boldsymbol{\xi}\| \leq 2R} |\widehat{\mu}(\boldsymbol{\xi})|^4 d\boldsymbol{\xi}. \tag{3.11}$$

By the Fubini theorem,

$$
\begin{aligned}
M_{E,4}(R) &\approx \int\int\left[\int e^{-2\pi i \boldsymbol{z}\cdot\boldsymbol{\xi}}\psi(\boldsymbol{\xi}/R)d\boldsymbol{\xi}\right]d\mu_X d\mu_Y \\
&= R^n\int\int\widehat{\psi}(R\boldsymbol{z})d\mu_X d\mu_Y,
\end{aligned}
\tag{3.12}
$$

where $\boldsymbol{z} = \boldsymbol{x}_1 + \boldsymbol{x}_2 - \boldsymbol{y}_1 - \boldsymbol{y}_2$. Hence, since $\widehat{\psi}$ decays rapidly, we get

$$
M_{E,4}(R) \approx R^n\cdot\mu^*\{(\boldsymbol{x}_1,\boldsymbol{x}_2,\boldsymbol{y}_1,\boldsymbol{y}_2) : \boldsymbol{x}_1+\boldsymbol{x}_2 \approx_\delta \boldsymbol{y}_1+\boldsymbol{y}_2\},
\tag{3.13}
$$

where $\delta \approx \frac{1}{R}$.

Furthermore, the formula (3.1) vindicates the following discretization. Let $N \approx R$ be an integer. There exists a set $\Gamma_N \subset (NE\cap\mathbb{Z}^n)$ of cardinality $\approx N^{\frac{n}{2}}$, such that

$$
\begin{aligned}
&\mu^*\{(\boldsymbol{x}_1,\boldsymbol{x}_2,\boldsymbol{y}_1,\boldsymbol{y}_2) : \boldsymbol{x}_1+\boldsymbol{x}_2 \approx_\delta \boldsymbol{y}_1+\boldsymbol{y}_2\} \\
\approx\;& N^{-2n}\,\#\{(\boldsymbol{a}_1,\boldsymbol{a}_2,\boldsymbol{b}_1,\boldsymbol{b}_2)\in\Gamma_N : \boldsymbol{a}_1+\boldsymbol{a}_2=\boldsymbol{b}_1+\boldsymbol{b}_2\}.
\end{aligned}
\tag{3.14}
$$

Observe that the maximum number of solutions of the above discrete equation is $\lesssim N^{\frac{3n}{2}}$, as a specific triple $(\boldsymbol{a}_1,\boldsymbol{a}_2,\boldsymbol{b}_1)$ fixes $\boldsymbol{b}_2$.

In view of this, let us consider the case when the fourth moment is large. Namely assume that for all R large enough, one has

$$
M_{E,4}(R) \gtrsim R^{\frac{n}{2}}.
\tag{3.15}
$$

This condition is tantamount to saying that the discrete equation in (3.14) has the maximum order of magnitude for the number of solutions: $\gtrsim N^{\frac{3}{2}}$.

Then we can establish the principal result of this section; before we do this let us introduce some definitions.

- We say that a finite set $\mathbb{A}$ is an *arithmetic progression* in $\mathbb{Z}^n$ of dimension k and size L, if each element of $\mathfrak{g}\in\mathbb{A}$ possesses a representation

$$
\mathfrak{g} = \mathfrak{g}_0 + \{r_1\mathfrak{g}_1 + \cdots + r_k\mathfrak{g}_k\}_{1\le r_j\le L_j},
\tag{3.16}
$$

  where each r_j is an integer, each $\mathfrak{g}_j$ is a (fixed) element of $\mathbb{Z}^n$ (called a generator), and $L_1\cdot L_2\cdot\cdots\cdot L_k = L$. An arithmetic progression is *proper* if the representation (3.16) is unique for each $\mathfrak{g}\in\mathbb{A}$.

- Let us call a measure μ, satisfying (3.1) and supported on a compact set E *arithmetic* if there exists a positive measure subset $E'\subset E$, such that for each δ sufficiently small, E'_δ, the δ-neighborhood of E' is contained in the δ-neighborhood of a dilate of a proper arithmetic progression $\mathbb{A}=\mathbb{A}(\delta)$ in $\mathbb{Z}^n$. (If $\alpha>0$, the progression has to get longer as $\delta\to 0$.)

Theorem 3.1. *If (3.15) holds for all sufficiently large R, the measure μ is arithmetic.*

The condition (3.15) is in particular satisfied in the case $n = 2$, when E is the boundary of a polygon, which certainly satisfies Theorem 3.1.

Theorem 3.1 will follow from the following classical combinatorial theorem, see [6], [3].

Theorem 3.2 (Freiman's theorem). *Let A_N, $B_N \subset \mathbb{Z}^n$ such that $\#A_N = \#B_N = N$, and $\#(A_N + B_N) \le CN$, where C is independent of N and*

$$A_N + B_N = \{\boldsymbol{a} + \boldsymbol{b},\ \boldsymbol{a} \in A_N,\, \boldsymbol{b} \in B_N\}.$$

Then A_N is contained in some k-dimensional arithmetic progression in $\mathbb{Z}^n$, where k depends only on C.

Freiman's theorem can be applied to (3.14, 3.15) as follows. Consider the set

$$2\Gamma_N = \Gamma_N + \Gamma_N = \{\boldsymbol{u} = \boldsymbol{a} + \boldsymbol{b},\ \boldsymbol{a}, \boldsymbol{b} \in \Gamma_N\},$$

let $f(\boldsymbol{u})$ be the number of representations $\boldsymbol{u} = \boldsymbol{a} + \boldsymbol{b}$ for each $\boldsymbol{u} \in 2\Gamma_N$. Then the number of solutions of the discrete equation in (3.14) can be expressed as

$$\begin{aligned} N^{\frac{3}{2}} \lesssim \textstyle\sum_{\boldsymbol{u}\in 2\Gamma_N} f^2(\boldsymbol{u}) &\le \max_{\boldsymbol{u}\in 2\Gamma_N} f(\boldsymbol{u}) \cdot \textstyle\sum_{\boldsymbol{u}\in 2\Gamma_N} f(\boldsymbol{u}) \\ &\lesssim N^{\frac{n}{2}} \cdot N^{2\cdot\frac{n}{2}} = N^{\frac{3n}{2}}. \end{aligned} \tag{3.17}$$

From this estimate we conclude that there is a subset $\Upsilon_N \subset 2\Gamma_N$ of cardinality $\approx N^{\frac{n}{2}}$, such that for all $\boldsymbol{u} \in \Upsilon_N$ we have $f(\boldsymbol{u}) \gtrsim N^{\frac{n}{2}}$. Indeed,

$$\#\Upsilon_N \lesssim \frac{N^n}{N^{\frac{n}{2}}} = N^{\frac{n}{2}} \lesssim \#\Gamma_N. \tag{3.18}$$

Then there exist subsets $\Gamma_{1,N}, \Gamma_{2,N}$, each of cardinality of $\approx \#\Gamma_N \approx N^{\frac{n}{2}}$, such that

$$\Gamma_{1,N} + \Gamma_{2,N} = \Upsilon_N. \tag{3.19}$$

Besides, each set $\Gamma_{j,N}$ cannot have a diameter $o(N)$, as this would contradict (3.1): one cannot have a positive proportion of the elements of Γ_N inside a set whose volume is $o(1)$. This proves Theorem 3.1 and also implies that the set Γ_N contains a straight line on which there sit $\gtrsim N$ of its members. This also vindicates the Falconer conjecture for sets, such that (3.15) is satisfied, i.e. sets that have a lot of additive structure.

In conclusion, let us consider the opposite case to a strong additive structure on E. Namely, we say that a measure μ on E is *strictly convex* if the equation

$$\boldsymbol{x} + \boldsymbol{y} = \boldsymbol{x}' + \boldsymbol{y}', \quad \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{x}', \boldsymbol{y}' \in E \tag{3.20}$$

has an at most bounded number of non-trivial solutions, i.e. those when $\boldsymbol{x}$ is not equal $\boldsymbol{x}'$ or $\boldsymbol{y}'$.

Let us show that the Falconer conjecture holds for strictly convex measures. That is we have to prove that $M_{E,2} \lesssim 1$. By the Minkowski integral inequality

$$\left(\int \left(\int K^p(\boldsymbol{x},\boldsymbol{y})d\boldsymbol{x}\right)^{\frac{q}{p}} d\boldsymbol{y}\right)^{\frac{1}{q}} \leq \int \left(\int K^q(\boldsymbol{x},\boldsymbol{y})d\boldsymbol{y}\right)^{\frac{1}{q}} d\boldsymbol{x}, \text{ for } 1 < p < q,$$

we have

$$\begin{aligned} M_{E,2} \leq M_{E,4} &\lesssim R^n \, \mu^*\{(\boldsymbol{x},\boldsymbol{y},\boldsymbol{x}',\boldsymbol{y}') : \boldsymbol{x}+\boldsymbol{y} =_{R^{-1}} \boldsymbol{x}'+\boldsymbol{y}'\}, \\ &\lesssim R^{n-2\alpha} \lesssim 1, \end{aligned} \tag{3.21}$$

for $\alpha \geq \frac{n}{2}$, as desired. We conclude that an "anti-additive" case, i.e. when the discrete equation in (3.14) has the minimum number of solutions, the fourth moment $M_{E,4}$ is extremely small. The Falconer distance problem gets vindicated in both cases however. The real problem in this respect is what happens in between.

From the material fingerprinting point of view, the two aspects considered in this section indicate that the pair of moments $M_{E,2}$ and $M_{E,4}$ represent important information regarding the distance and addition structure of the sample set E, and therefore are likely to identify promptly different types of translational symmetry of crystal matter.

In conclusion, clearly the same can be said about higher moments $M_{E,2l}$, $l > 2$, which would yield the discrete equation

$$\boldsymbol{a}_1 + \ldots + \boldsymbol{a}_l = \boldsymbol{b}_1 + \ldots + \boldsymbol{b}_l,$$

cf. (3.14). In particular, Theorem 3.1 applies to higher moments with the same estimate (3.15). In view of this we conjecture that an array of moments $\{M_{E,2l}\}_{l\geq 1}$ should represent an unambiguous signature of a given material.

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