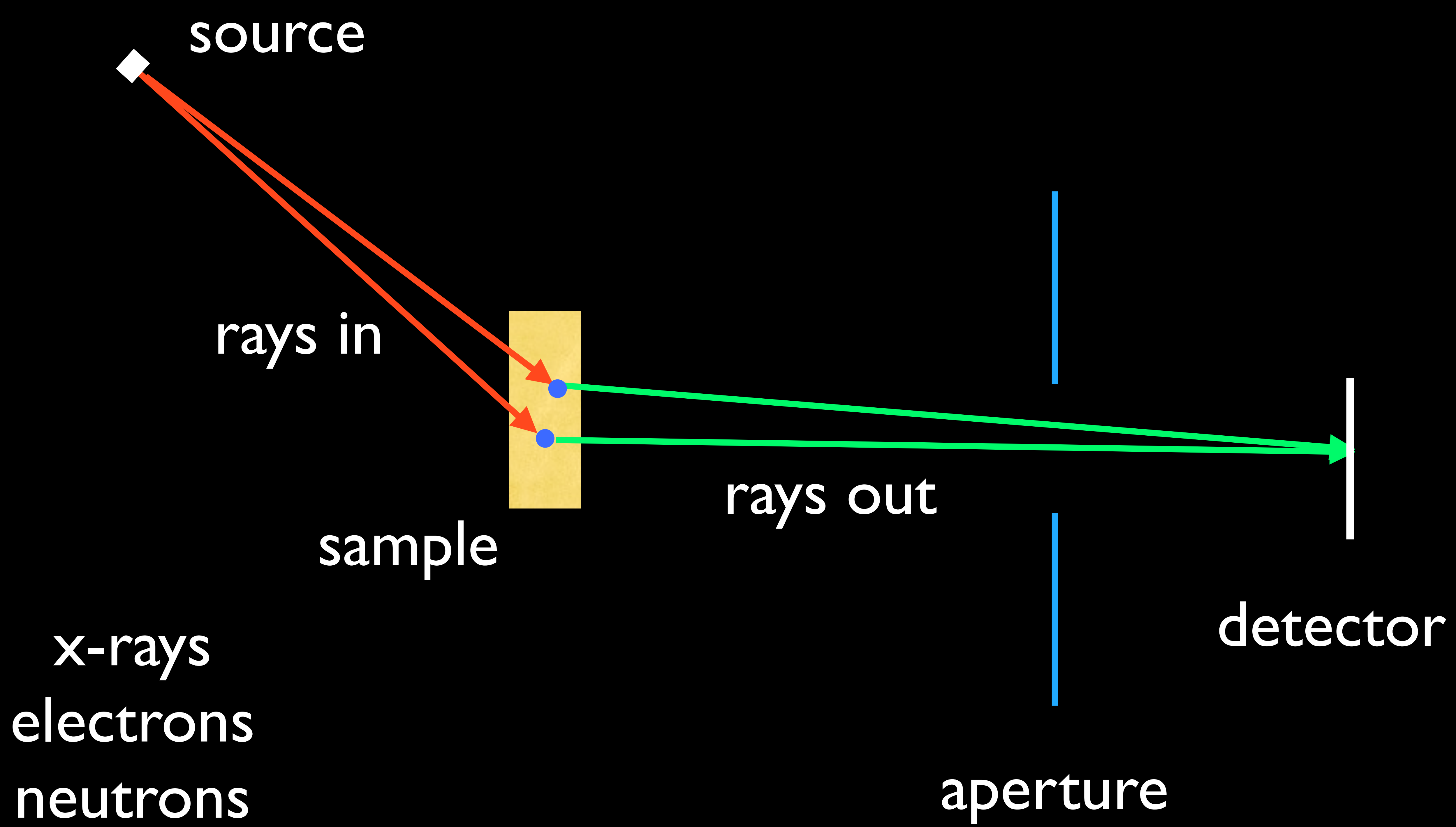
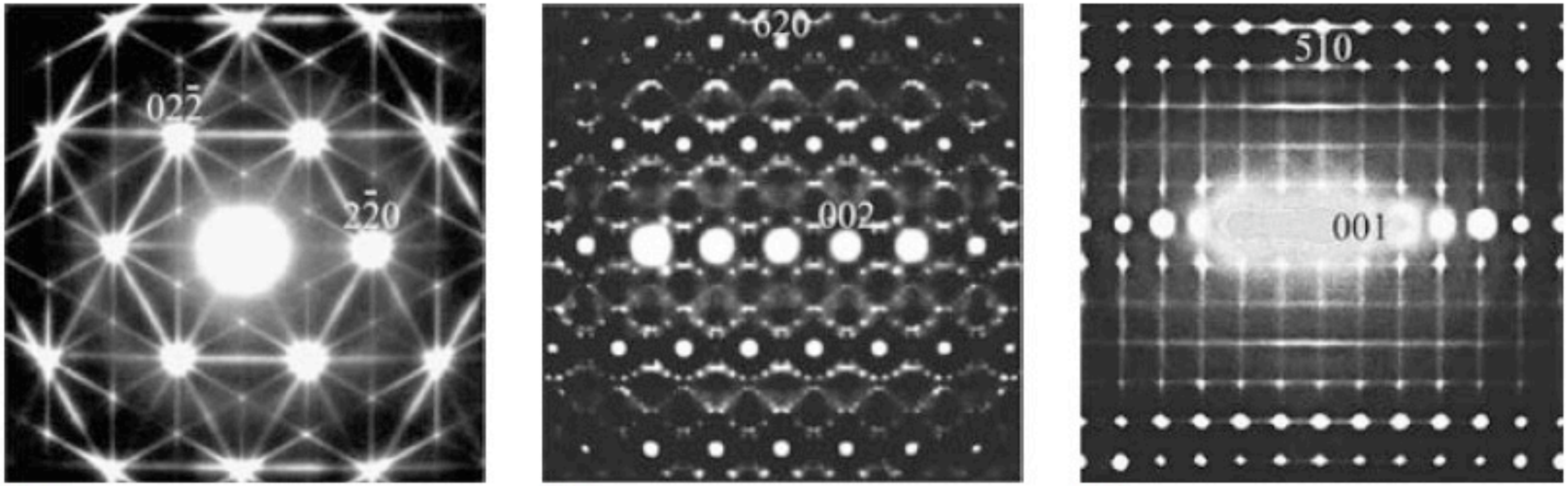
Stationary spatial stochastic processes and the inverse problem in pure point diffraction

Robert V. Moody, University of Victoria

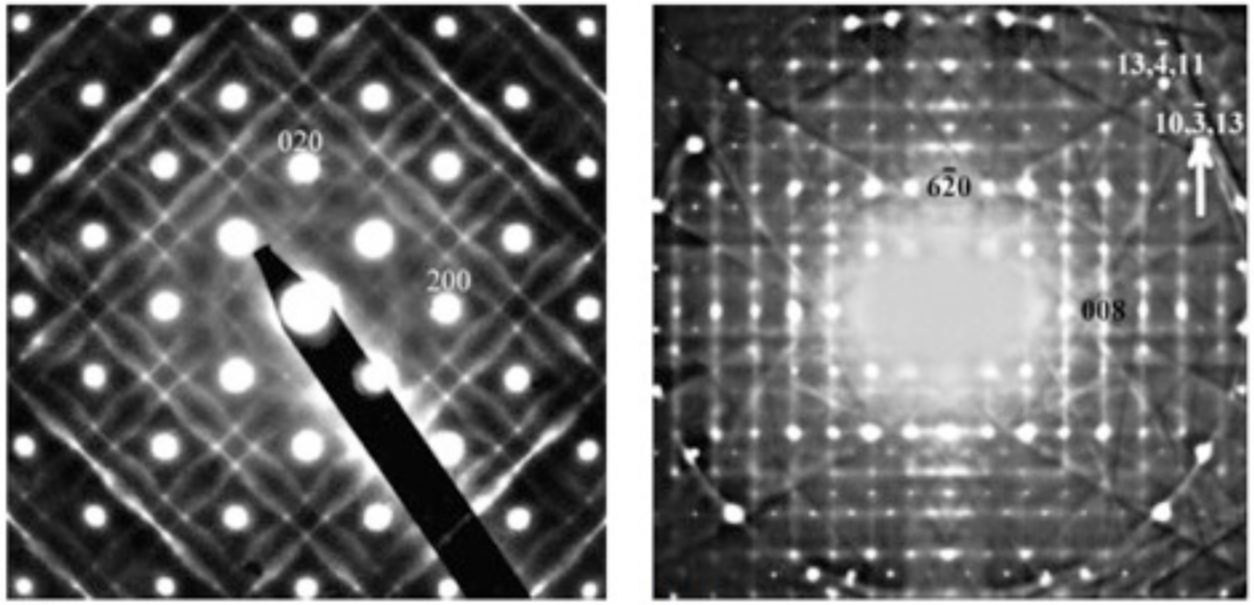
BIRS, September 2011



Microscope/Diffractometer



(a) (b) (c)



(a) (b)

FIGURE 4 An (a) [001] zone axis EDP of *P4/nmm* ThAsSe (see Withers *et al.*, 2006 for details) and (b) a $\langle 130 \rangle$ zone axis electron diffraction pattern of *Im* − 3 CCTO (see Liu *et al.*, 2005 for details). Note that the $\mathbf{G} \pm \sim 0.14 \langle 110 \rangle^* \pm \varepsilon \langle 1, -1, 0 \rangle^*$ diffuse streaking in (a) occurs only around the $h + k$ odd, *n*-glide forbidden parent reflections, whereas the diffuse streaking perpendicular to $\langle 001 \rangle$ in (b) only runs through the $[hkl]^*$, l even parent reflections.

from R. Withers: Disorder: structured diffuse scattering and local crystal chemistry

Diffraction

self-interference of scattered radiation from a scattering density or distribution of matter ρ

ρ

- could be a set of point masses
- the electric field of an assemblage of atoms
- more generally a measure in $\mathbb{R}^d$

Mathematically the diffraction is

$$\omega = |\widehat{\rho}|^2$$

treated as a measure in $\mathbb{R}^d$

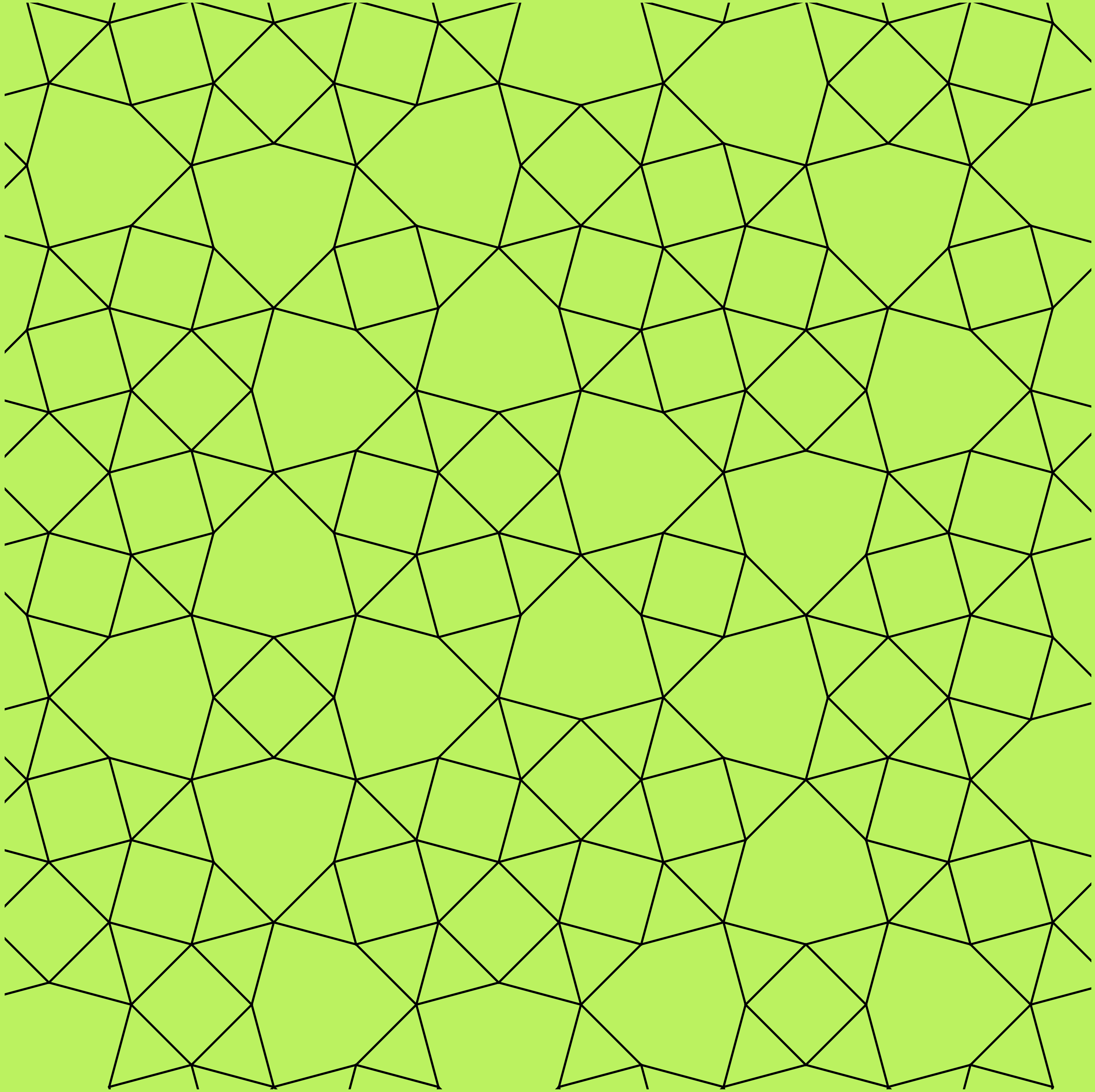
The diffraction is a measure in reciprocal space -- the Fourier dual of the physical space

In the case of $\mathbb{R}^d$ this is another copy of $\mathbb{R}^d$.

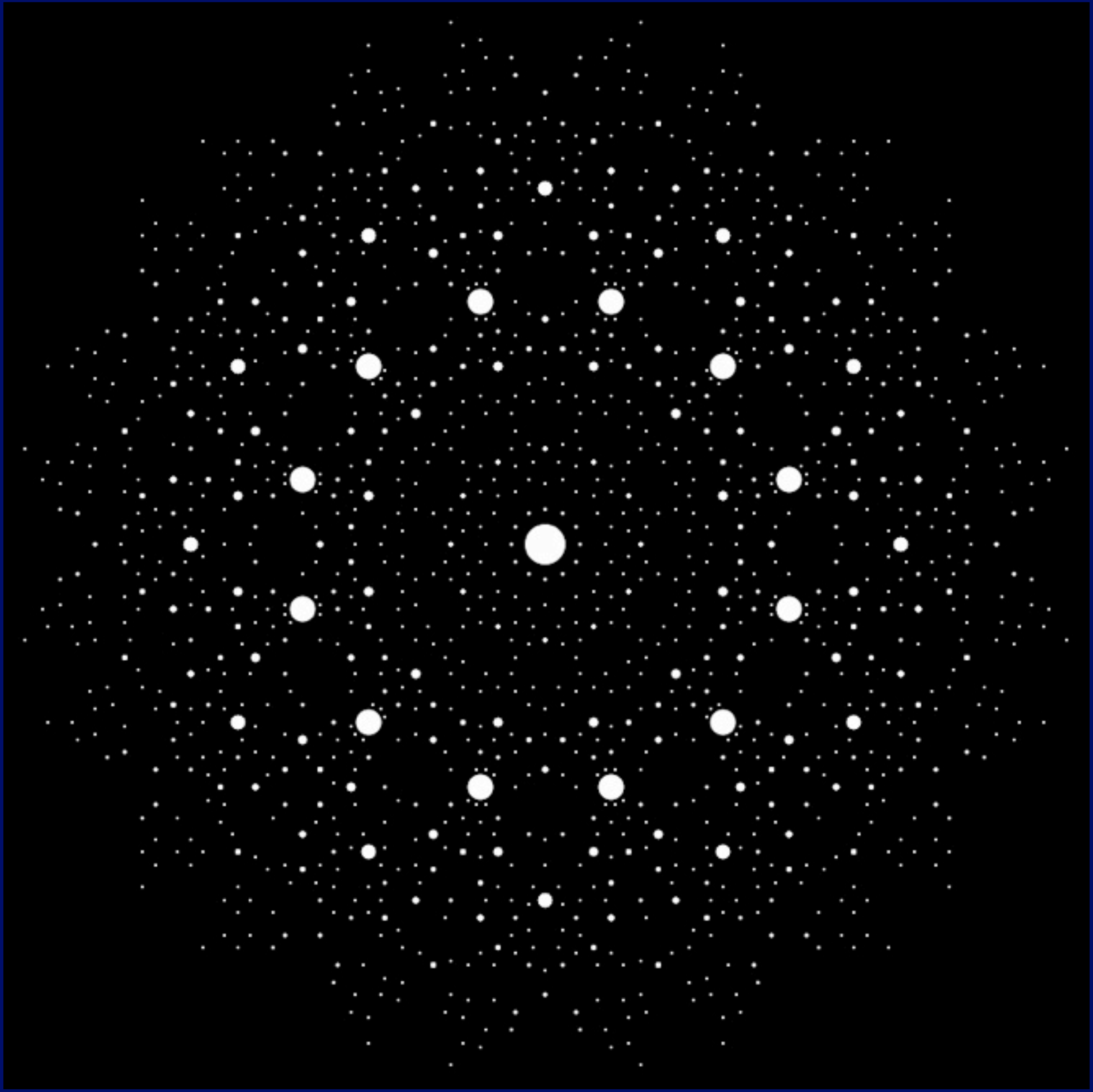
Later $\mathbb{R}^d$ will be replaced by any locally compact Abelian group G, Fourier space becomes $\widehat{G}$.

Three basic properties of the diffraction measure

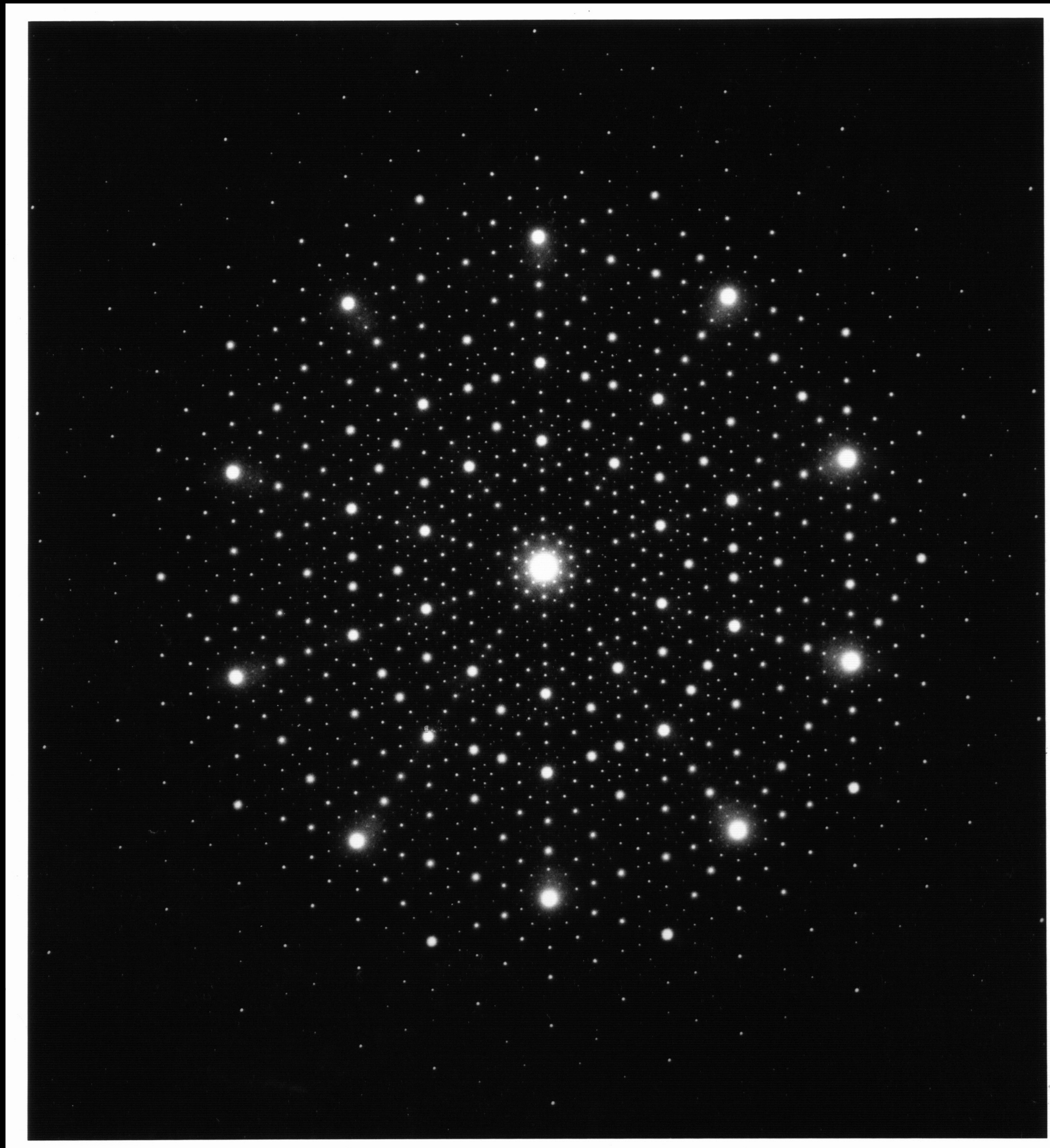
- It is positive
- It is centrally symmetric
- It is a translation bounded measure



The Shield Tiling -- Franz Gaehler



Diffraction of the Shield Tiling



the diffraction is a positive and centrally symmetric measure in Fourier space

The central problem of diffraction theory:

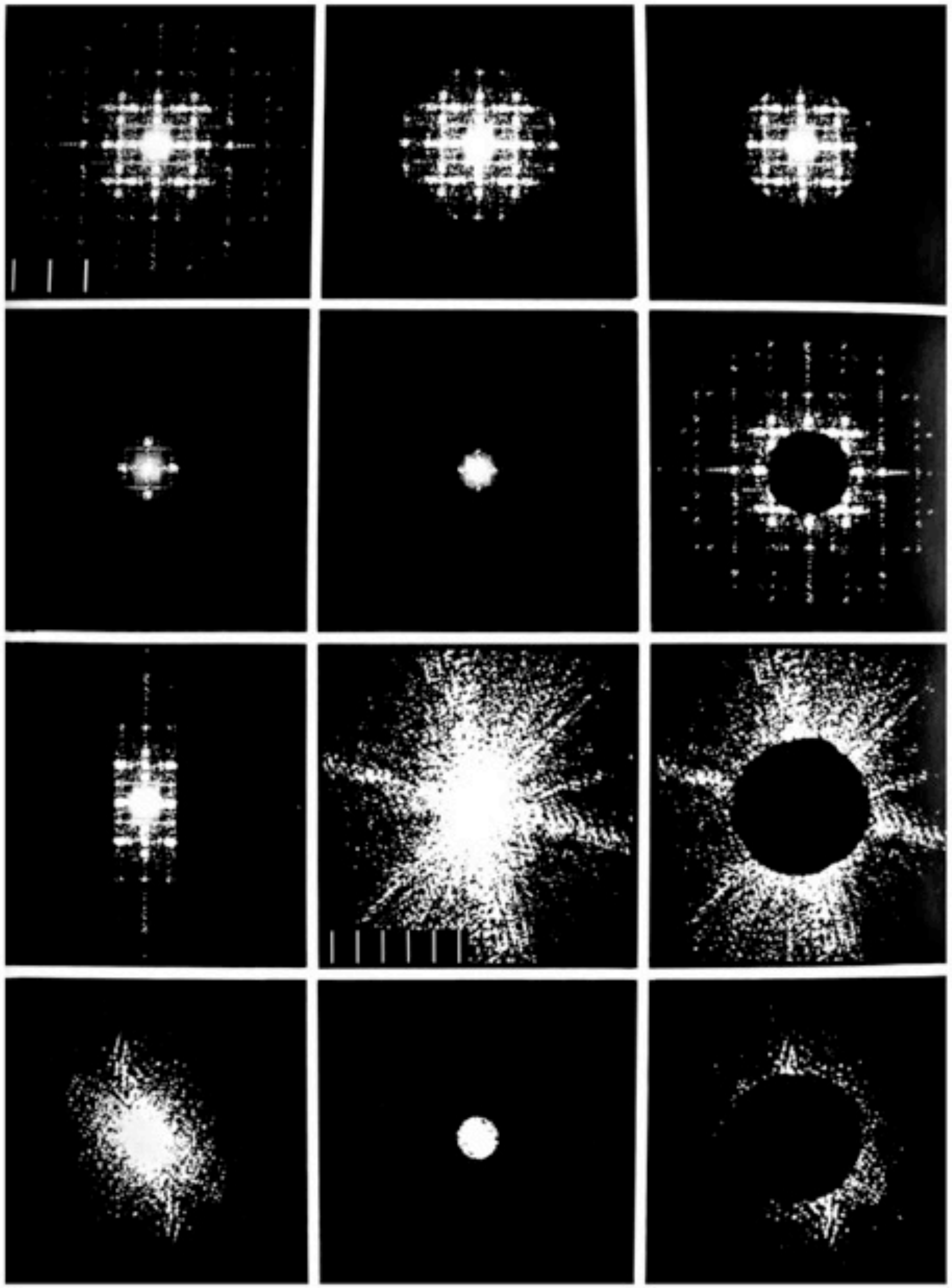
given the diffraction, determine the structure that created it

Why it is hard: We want ρ

We are given $\omega = |\widehat{\rho}|^2$ This is the Phase Problem

$$\begin{aligned} \omega &= \widehat{\rho}\,\overline{\widehat{\rho}} \\ &= \widehat{\rho * \tilde{\rho}} = \widehat{\gamma} \end{aligned}$$

the Patterson "function" a.k.a. the autocorrelation



Brief History

Knipping/Laue experiment 1912

Bragg and Bragg (William Henry and William Lawrence)
1913 first explanations

Arthur (Lindo) Patterson function 1934

Linus Pauling 1939 bixbyite – homometry

D. Shechtman, quasicrystals, 1982

different structures
with the same
diffraction

Three problems

1. Given a centrally symmetric positive translation bounded measure, find a structure which has it as its diffraction.

2. Classify all the structures with the same diffraction.

3. What do we mean by a “structure”?

In the pure point case we can give reasonable answers to these three questions

A key point is to introduce the notion of stationary stochastic processes

This is the main subject of this talk

joint work with Daniel Lenz, Jena, Germany

stationary spatial stochastic process

$$\begin{aligned} N : C_c(\mathbb{R}^d) &\longrightarrow L^2(X,\mu) \\ F &\mapsto N(F) \end{aligned}$$

$\boxed{C_c(\mathbb{R}^d)}$

the space of continuous functions of compact support

$\boxed{(X,\mu) = (X, \mathbb{R}^d, \mu)}$

a measure dynamical system with an $\mathbb{R}^d$-action on it

$\boxed{\mu}$ is stationary (translation invariant)

$\boxed{N}$ is a linear $\mathbb{R}^d$ - map

$\boxed{N}$ is real: real valued functions go to real valued functions

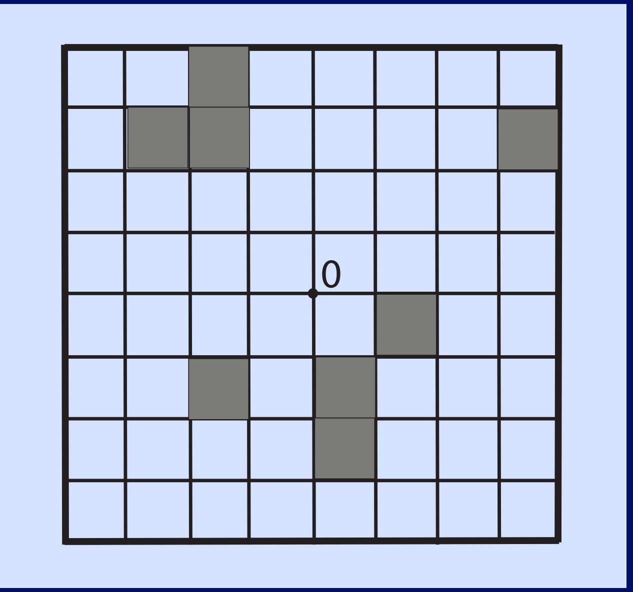
motivation from stochastic point processes

a random variable whose outcome for each event is a discrete point set in $\mathbb{R}^d$

X is the set of all possible outcomes.

each $\xi \in X$ is a discrete point set in $\mathbb{R}^d$.

X is assumed to be $\mathbb{R}^d$-invariant



measurable sets

given a pattern , Π

$B(\Pi) =$ all $\Lambda \in X$ matching the pattern Π

these generate our sigma-algbebra of measurable sets

X is a stationary probability space

probability, or frequency measure μ

$$\mu(B(\Pi)) = \text{frequency of occurrence of the pattern } \Pi$$

$$\begin{cases} \mu \text{ is translation invariant (stationary)} \\ \mu(X) = 1 \end{cases}$$

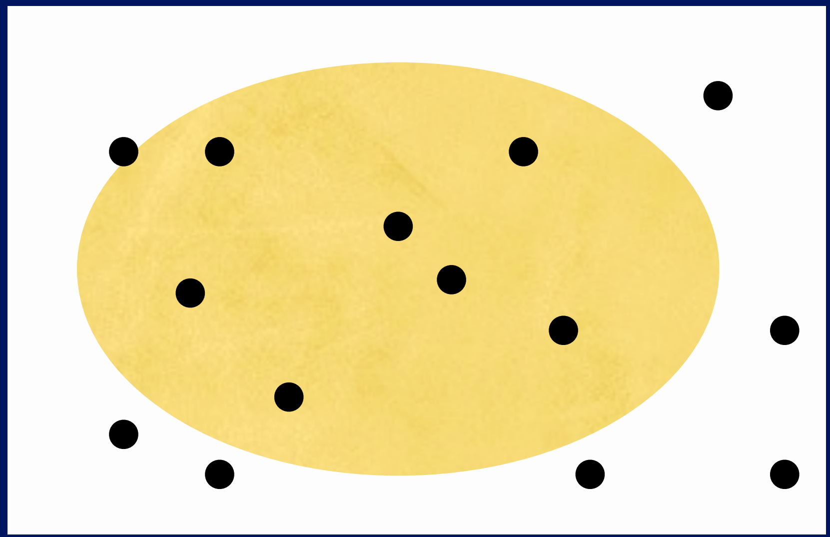
$$\Longrightarrow \quad (X, \mathbb{R}^d, \mu)$$

probability space
measure theoretic dynamical system

Assume the system is ergodic

defining N

For each measurable set $A \subset \mathbb{R}^d$ and for each $\xi \in X$



$$N_A(\xi) = \text{the number of points of } \xi \text{ in } A$$
$$= \sum_{x \in \xi} 1_A(x)$$

$$N_A : X \longrightarrow \mathbb{R}$$

counting functions

In effect, N provides a link between the physical space of the objects which are labelled by the elements of X

extend to functions

F function on $\mathbb{R}^d$ $\longrightarrow$ $N(F)$ function on X

$$N(F)(\xi) = \sum_{x \in \xi} F(x)$$

$$N : C_c(\mathbb{R}^d) \longrightarrow L^2(X, \mu)$$

stationary spatial stochastic processes (sssp)

We shall always assume that N has a second moment (to come soon!)

$$(X,\ \mathbb{R}^d,\ \mu,\ N)$$

the hull: compact space

physical space

probability measure

counting function

spatial stationary stochastic process

$$N : C_c(\mathbb{R}^d) \longrightarrow L^2(X,\mu)$$

$$F \mapsto N(F)$$

how to think about N

$$N : C_c(\mathbb{R}^d) \longrightarrow L^2(X, \mu)$$

$$F \mapsto N(F)$$

Think of N as providing a way of providing test functions for the various elements of X

$$\xi : F \mapsto N(F)(\xi) \text{ for a.e. } \xi \in X.$$

Comment

G locally compact Abelian group

$\mathcal{H}$ Hilbert space

$$N : C_c(\cancel{\mathbb{R}^d}) \longrightarrow L^2(\cancel{X}, \mu)$$

$$F \mapsto N(F)$$

spatial versus non-spatial

two types: sssp and ssp

Second moments

$$N \text{ has a second moment if}$$
$$\text{for all real-valued } F, G \in C_c(\mathbb{R}^d)$$

$$\mu^{(2)}(F \otimes G) = \langle N(F) | N(G) \rangle$$

$$\text{is a measure on } C_c(\mathbb{R}^d \times \mathbb{R}^d)$$

$\longrightarrow$

there exists a positive definite measure γ on $\mathbb{R}^d$ so that

$$\int_{\mathbb{R}^d} F * \tilde{F} \, d\gamma = \langle N(F) | N(F) \rangle$$

γ is the autocorrelation

Its Fourier transform $\omega = \widehat{\gamma}$ is the diffraction

ω is positive, centrally symmetric, and translation bounded.

The question: given

ω is positive, centrally symmetric, and translation bounded.

Does there exist a stationary spatial stochastic process with this as its diffraction?

Yes, in the pure point case

pure point diffraction means ω is a pure point measure.

Theorem:

In the ergodic case this is equivalent to $L^2(X, \mu)$ is a pure point dynamical system

Some results

N has a second moment iff
for all real-valued $F, G \in C_c(\mathbb{R}^d)$
with support in the compact set K
there is a constant C_K so that
$\langle N(F)|N(G)\rangle \leq C_K \max\{||F||_2 \, ||G||_2, ||F||_\infty \, ||G||_1\}$

Any stationary spatial stochastic process N(sssp)
with diffraction ω splits into $N = N_p + N_c$ where
N_p is a pure point sssp with diffraction ω_p
N_c is a continuous ssp with diffraction ω_c.

A sssp N has pure point diffraction if and only if
for all $F \in C_c(\mathbb{R}^d)$, $t \mapsto T_t \, N(F)$
is strongly almost periodic with respect to the
L^2 norm on $L^2(X, \mu)$.

How does one construct a sssp from a given pure point positive centrally symmetric translation bounded measure?

Start with $\omega = \sum_{k \in S} \omega(k)\, \delta_k$

S is the set of Bragg peaks

There are two ways to approach this

Both need the concept of a phase form

$$a : S \longrightarrow U(1) \text{ (the unit circle in } \mathbb{C})$$

$$a(0) = 1$$

$$a(-k) = \overline{a(k)} \qquad \text{for all } k \in S.$$

1. Since $L(X, \mu)$ should be pure point,
Halmos-von Neumann says that measure theoretically
X is a compact Abelian group.
$E := \langle S \rangle \subset \mathbb{R}^d$
is the subgroup generated by the Bragg peaks.

Give it the discrete topology and put
$X = \widehat{E}$.

This is a compact Abelian group
and with usual Haar measure serves as (X, μ).

the phase form enters in the definition of N

2. Use Gel'fand theory

form a commutative Banach algebra
and make X its set of maximal ideals

for $k \in S$, put $c(k) = a(k)\,\omega(k)^{1/2}$

$\mathcal{P}$ is defined as the free commuative algebra over $\mathbb{C}$
with generators f_k, $k \in S$
relations $f_{k_1} \cdots f_{k_m} = c(k_1) \cdots c(k_m)$
whenever $k_1 + \cdots + k_m = 0.$

action of $\mathbb{R}^d$

$t.f_k = (k,t) f_k$

inner product

$$\langle f_{k_1} \cdots f_{k_m} | f_{l_1} \cdots f_{l_n} \rangle = \begin{cases} c(k_1) \cdots c(k_m) c(-l_1) \cdots c(-l_n) \\ 0 \end{cases}$$

according as $k_1 + \cdots + k_m = l_1 + \cdots + l_n$ or not.

Use this to define the operator norm ν on $\mathbb{P}$:
$\nu(x)$ is the norm of multiplication by x.

$\mathcal{A}$ is the completion of $\mathcal{P}$ under ν.

$\mathcal{A}$ is a commutative Banach algebra

X is its maximal ideal spectrum.

skip the definition of the probability measure μ

$$\begin{aligned} N : C_c(\mathbb{R}^d) &\longrightarrow L^2(x, \mu) \\ F &\mapsto \sum_{k \in S} \widehat{F}(k) f_k \end{aligned}$$

this is the desired sssp with diffraction ω.

$$(X,\ \mathbb{R}^d,\ \mu,\ N)$$

the hull: compact space

physical space

probability measure

counting function

spatial stationary stochastic process

$$N : C_c(\mathbb{R}^d) \longrightarrow L^2(X,\mu)$$
$$F \mapsto N(F)$$

The remaining problem is to classify phase forms and decide when they lead to equivalent sssp's

But what does the density really look like?

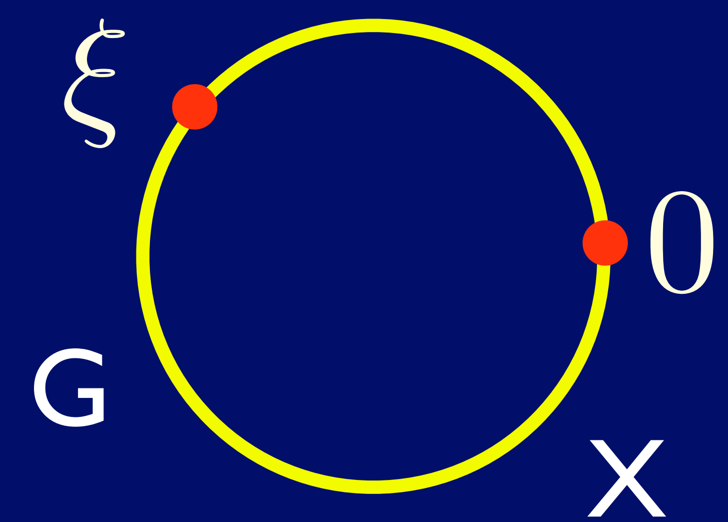
Example

periodic structure on $\mathbb{R}$ with diffraction $\sum_{k\in\mathbb{Z}} \delta_k$

$$G = U(1) \qquad \widehat{G} = \mathbb{Z} \qquad X \simeq U(1)$$

$$\omega = \sum_{k\in\mathbb{Z}} \delta_k$$

$$\begin{aligned} N_a(F) &= \sum_{k\in\mathbb{Z}} \widehat{F}(k)\, a(k)\, \chi_k \\ &= \sum_{k\in\mathbb{Z}} \widehat{F}(k)\, a(k)\, e^{2\pi i k.(\cdot)} \end{aligned}$$



If $a \equiv 1$ then

$$\begin{aligned} N_1(F)(\xi) &= \sum_{k\in\mathbb{Z}} \widehat{F}(k)\, \chi_k(\xi) \\ &= \sum_{k\in\mathbb{Z}} \widehat{F}(k)\, e^{2\pi i k.(\xi)} = F(\xi) = \delta_\xi(F) \end{aligned}$$

$$\boxed{N_1 \Leftrightarrow \rho = \delta_0}$$

What does it mean that $N_1(F(\xi) = \delta_\xi(F)$?

N_1 is supposed to offer some sort of 'density' at each point $\xi_0 \in X$.

$F \mapsto N_1(F)(\xi)$ shows how test functions interact with the density

In our case, X is just one orbit – that of $1 \in U(1)$.

From translation invariance of N
we expect
density at ξ = translation by ξ of the density at 0.

$$N_1 \leftrightarrow \delta_0$$

Example continued

Fix a finite set $K = -K \subset \mathbb{Z} \backslash \{0\}$.

Define phase form a by

$$a(k) = \begin{cases} 1, & \text{if } k \notin K \\ \text{arbitrary in } U(1), & \text{otherwise}. \end{cases}$$

with $a(-k) = \overline{a(k)}$.

Then $\rho = \delta_0 + \sum_{k \in K} (a(k) - 1) e^{-2\pi i k.(\cdot)}$.

Remarkably this has the same diffraction, $\omega = \sum_{k \in \mathbb{Z}} \delta_k$

Moments

the m th moment

$$\mu^{(m)}(F_1 \otimes \cdots \otimes F_m) = \int N(F_1) \dots N(F_m) d\mu$$

the second moment determines diffraction

knowing all moments determines μ

Example (Grunbaum and Moore)

$$G = \mathbb{Z}/6\mathbb{Z}, \quad \widehat{G} = \tfrac{1}{6}\mathbb{Z}/\mathbb{Z}$$

$$\rho_1 = 11\delta_0 + 25\delta_1 + 42\delta_2 + 45\delta_3 + 31\delta_4 + 14\delta_5$$

$$\rho_2 = 10\delta_0 + 17\delta_1 + 35\delta_2 + 46\delta_3 + 39\delta_4 + 21\delta_5$$

these two densities have the same their first 5 moments equal!!.

Summary

We can explicitly define sssp for any pure point measure that looks like a diffraction measure

We can classify all such sssp's

The notion of an sssp is very general and offers a good context for studying diffraction

The most outstanding problem is that we do not know if sssp's are always due to some sort of measure on the physical space.

We have no constructive theory for continuous or mixed diffraction types

Charles Radin
Stephen Dworkin
Bert Hof
Martin Schlottmann
Jean Bellissard
Jean-Baptiste Gouéré
Michael Baake
Daniel Lenz
Nicolae Strungaru