A GLIMPSE AT MATHEMATICAL DIFFRACTION THEORY

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Mathematical diffraction theory is concerned with the analysis of the diffraction measure of a translation bounded complex measure ω. It emerges as the Fourier transform of the autocorrelation measure of ω. The mathematically rigorous approach has produced a number of interesting results in the context of perfect and random systems, some of which are summarized here.

1 Introduction

Diffraction experiments are widely used to gain information on the atomic structure of solids. Neutron and X-ray diffraction share the property that fundamental aspects of the diffraction image can be understood very well in single scattering theory together with Born's approximation, while electron diffraction needs more sophistication, see Ref. 7 for details. In the former case, on which we will focus here, the diffraction image is then described as the Fourier transform of the autocorrelation of the original distribution of scatterers. In this situation, important information is lost, namely the relative phases. Consequently, the inverse problem of structure determination has, in general, no unique solution, unless extra information is available from other sources of knowledge.

All this can be phrased in mathematically rigorous terms, but hardly anybody seemed really interested in it – until quasicrystals were discovered in the early 1980's. They are solids which, like crystals, show sharp Bragg diffraction images in X-ray diffraction, yet with maliciously perfect point symmetries which are crystallographically forbidden – at least in dimensions ≤ 3. As turned out quickly, they can be formally described as projections of portions (strips) of lattices in higher dimensions, and this can account both for their symmetries and for their perfect Bragg diffraction spectra. However, they also leave a certain degree of uneasiness towards the obviously not sufficiently well understood possibilities in diffraction theory.

This was the starting point of the rigorous development of mathematical diffraction theory in a paper by Hof [9] which was later also extended to the treatment of high temperature.[10] The setting is as follows. The structure under investigation is described as a translation bounded complex measure ω

on n-dimensional Euclidean space, whose autocorrelation, γ_ω is obtained as a volume-weighted convolution,

$$\gamma_\omega \;=\; \lim_{r\to\infty} \frac{\omega|_r * \tilde{\omega}|_r}{\mathrm{vol}(B_r)} \tag{1}$$

where B_r is the (open) ball of radius r around 0, $\omega|_r$ denotes the restriction of ω to B_r, and $\tilde{}$ stands for taking the complex conjugate of the origin-inverted measure. It is not even clear in general whether this limit exists, but if not, the right hand side certainly has points of accumulation which can then be obtained as limits along a discrete subsequence of averaging balls. We will thus assume, from now on, that the limit in (1) exists.

If so, the autocorrelation is a positive definite measure, and as such automatically possesses a Fourier transform, denoted by $\hat{\gamma}_\omega$, which is a positive measure by Bochner's theorem, compare Ref. 6 for background material. Furthermore, $\hat{\gamma}_\omega$ has a unique decomposition into three parts,

$$\hat{\gamma}_\omega \;=\; \bigl(\hat{\gamma}_\omega\bigr)_{pp} + \bigl(\hat{\gamma}_\omega\bigr)_{sc} + \bigl(\hat{\gamma}_\omega\bigr)_{ac}\,, \tag{2}$$

where *pp*, *sc* and *ac* as usual stand for pure point, singular continuous and absolutely continuous, with the ordinary Lebesgue measure as reference. The pure point (absolutely continuous) part corresponds to the Bragg spectrum (to diffuse scattering), while the term 'singular continuous' does not appear in the standard crystallographic literature.

In what follows, we will summarize a few aspects of this setting and give some pointers for further reading.

2 Pure point diffraction

One important class of structures are those with pure Bragg spectrum, i.e. those where $\hat{\gamma}_\omega$ is a pure point measure. This is certainly the case if the measure ω is crystallographic, i.e. periodic with a full lattice Γ of periods. So, $\omega = h * \delta_\Gamma$ where h is the density (or measure) in a fundamental domain and δ_Γ is the uniform lattice Dirac comb. With this, the autocorrelation reads $\gamma_\omega = (h * \tilde{h})\,\mathrm{dens}(\Gamma)\,\delta_\Gamma$, where $\mathrm{dens}(\Gamma)$ is the density of the lattice, and the diffraction measure is

$$\hat{\gamma}_\omega \;=\; |\hat{h}|^2 \,\bigl(\mathrm{dens}(\Gamma)\bigr)^2\, \delta_{\Gamma^*} \tag{3}$$

with $\Gamma^* = \{u \mid uv \in \mathbb{Z} \text{ for all } v \in \Gamma\}$ the dual lattice. This rests upon Poisson's summation formula, $\hat{\delta}_\Gamma = \mathrm{dens}(\Gamma)\delta_{\Gamma^*}$, and reconfirms that the diffraction spectrum of a perfect crystal is pure point.

Other by now well-known examples of pure point diffractive systems include perfect quasicrystals as described by the projection method. More precisely, given a cut and project scheme,[14] a generic, regular model set based on it will produce a pure point diffraction spectrum. Here, a subset of a lattice is projected which lies in an incommensurately oriented strip with constant cross section. The latter is called window and lives in the so-called internal space, while the complement, which the pattern is projected to, is termed direct or physical space. The diffraction result was first proved by Hof [9] for Euclidean internal spaces with polytopal windows. It was later extended by various people to include more general internal spaces, and finally proved in full generality by Schlottmann.[15] This fuelled the speculation that basically only model sets have this property, but the set of visible lattice points [5] proved otherwise. In fact, model sets only form a relatively sparse set of examples with pure point diffraction.

After some effort into solving this puzzle, the answer uses a different box of tools and relates diffraction properties to almost periodicity of the autocorrelation measure. Partly based on results of Gil de Lamadrid and Argabright, it is shown in Ref. 4 that a translation bounded measure ω has pure point diffraction if and only if its autocorrelation γ_ω is strongly almost periodic as a measure. This criterion can actually be checked in many cases, and it also provides an independent proof of Schlottmann's theorem as a by-product. The result also shows that the diffraction problem really consists of two steps: one from ω to its autocorrelation, γ_ω, and one from there to the Fourier transform, $\hat{\gamma}_\omega$. The second step is ultimately easy because it is at least one-to-one, even though many open questions are still to be settled. So, let us take a look at the first step, which lies at the heart of the phase reconstruction problem.

3 The homometry problem

Two different measures can share the same autocorrelation, as is obvious from the limit in (1), in which case they are called *homometric*. In particular, modifying a given measure by a *finite* measure has no effect on the limit, i.e. on γ_ω. But much worse operations are possible, which makes the equivalence relation defined by homometry a rather nasty one. To show how bad the situation is, we recall a result from Ref. 8. There, the binary Rudin-Shapiro sequence, which is based on a substitution rule and thus has entropy 0, is realized as a Dirac comb along the integers. It has the same autocorrelation as the generic Bernoulli Dirac comb with occupation probability 1/2 per site, which is truly random and has maximal entropy, $\log(2)$. The homometry class also contains an example for each intermediate value of the entropy. This shows how bad the inverse problem really is, unless further information is available about the structure, or a general principle is invoked to select a 'reasonable' representative from the homometry class, e.g. by maximizing the entropy.

Another example is discussed in Ref. 1. A subset S of a lattice is homometric with its complement set, if both have the same density – no matter whether the subset itself is periodic, aperiodic or random. So, one can in particular construct homometric pairs for all possible types of spectra, pure and mixed. Note that the pure point part which is due to the underlying lattice can be made extinct by simply subtracting a multiple of the uniform lattice Dirac comb. This way, one can even realize purely singular continuous spectra, e.g. based upon substitution rules of Thue-Morse type.

4 Diffraction of lattice subsets

A subset of a lattice Γ inherits a good deal of the lattice structure, no matter how unusual the selection of lattice points may be. In particular, if we once again make the (rather weak) assumption that a given subset S has a unique autocorrelation, the diffraction measure $\hat{\gamma}_\omega$ of the Dirac comb δ_S is periodic, with the dual lattice Γ^* as lattice of periods.[1] What is more, the autocorrelation admits a representation in the form $\gamma_\omega = \Phi \cdot \delta_\Gamma$ with a Lipschitz continuous function Φ which interpolates the autocorrelation coefficients between the discrete lattice points. It can actually be chosen to have an extension to an entire function, which is a bit surprising.

In connection with this, the diffraction measure takes the form

$$\hat{\gamma}_\omega \;=\; \varrho * \delta_{\Gamma^*} \tag{4}$$

where ϱ is a finite, positive measure concentrated on a true fundamental domain of the dual lattice, see Ref. 1 for details. This means that the analysis of the spectral type can be done with ϱ, which makes it considerably easier.

This result, like many mentioned so far, can be generalized substantially, e.g. to weighted lattice subsets or to the setting of lattices in locally compact Abelian groups that are σ-compact. On the other hand, it has a wide range of applications. First of all, the above mentioned set of visible lattice points falls into the class, so must have a diffraction of the convolution form (4), as was previously derived explicitly.[5] Another class of interesting point sets are those obtained from lattice substitution systems which have recently received a lot of attention.[13] Finally, all lattice gases, with or without interaction, are covered. This gives a unified explanation for the periodicity observed in the diffraction of these stochastic systems.

5 Diffraction from stochastic point sets

Let us finally take a closer look at systems with some degree of randomness, either due to a lattice gas structure or due to an underlying random tiling approach. Bernoulli subsets of lattices and model sets are well understood, even in terms of elementary methods from stochastics, see Ref. 3 and references therein. Recently, this approach has been generalized considerably, with more sophisticated machinery, to cover stochastic selections from rather general Delone sets.[11] The outcome proves the folklore claim that uncorrelated random removal of scatterers has two effects, namely reducing the overall intensity of the diffraction of the fully occupied set, without changing the relative intensities, and adding a white noise type constant diffuse background.

More interesting would be a detailed understanding of the spectra of stochastic systems with interactions. Though some qualitative features are known, quantitative results are rare, i.e. basically restricted to systems in one dimension or to systems that can be mapped to exactly solvable models of statistical mechanics, see Ref. 2 and the literature given there. A particularly interesting question concerns the precise spectral nature of random tilings of the plane, both with crystallographic and non-crystallographic symmetries (which is essentially meant as symmetry on average here). While typical dimer based lattice random tilings have mixed spectra with pure point and absolutely continuous components,[2] some quasicrystal related random tilings seem to have singular continuous parts. There are very convincing scaling arguments and numerical simulations in favour of this observation, but no proof is available at present. This is an open challenge, together with the investigation how robust and hence relevant such a singular continuous diffraction spectrum would be for real world crystallography.

Acknowledgments

This summary grew out of discussions and cooperations with a number of colleagues, in particular Moritz Höffe, Robert V. Moody and Martin Schlottmann, whose input is gratefully acknowledged. I thank Uwe Grimm for reading the manuscript and suggesting a number of improvements.

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