How should one define a (weak) crystal?

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Abstract. We compare two proposals for the study of positional long-range order: one in terms of the spectrum of the translation operator, the other in terms of the Fourier spectrum. We point out that only the first one allows for the consideration of molecular, as opposed to atomic, (weakly) periodic structures. We illustrate this point on the Thue-Morse system.

KEY WORDS: Positional long-range order; Fourier spectrum;
spectrum of the shift.

One of the fundamental questions of statistical mechanics is to describe ground states of systems of many interacting particles. It is an important problem to decide if, and to what extent, ground states are ordered, crystalline or otherwise [1, 2]. For the description of positional long-range order, which is necessary for the study of both crystals (periodic structures) and non-crystalline ordered structures like quasicrystals [3, 4], "turbulent" crystals [5], and "weakly periodic structures" [6] there have been two different main approaches in the literature, both in terms of spectral properties.

In the first one, which has been advocated within the infinite-volume approach to statistical mechanics [5, 7, 8, 9, 10, 11, 12, 13], one considers the spectrum of the Euclidean group acting as unitary operators on an appropriate Hilbert space. For classical models this is the $L^2(\mu)$ space for a translation-invariant Gibbs measure or ground state measure μ. To be more precise, we will discuss classical lattice gas models, in which every site of a lattice Z^d can be occupied by one of two (or more generally a finite number of) different particles (one may also think about them as different orientations of a spin variable). An infinite lattice configuration is an assignment of particles to lattice sites, that is an element of $\Omega = \{-1, +1\}^{Z^d}$. We will be mostly concerned with configurations which have uniformly defined frequencies for all finite patterns. More precisely, to find the frequency of a finite pattern in a given configuration we first count the number of times it appears in a box of size l and centered at the origin of the lattice, divide it by l^d, and then take the limit $l \to \infty$. If the convergence is uniform with respect to the position of the boxes then we say that the configuration has a uniformly defined frequency of this pattern. The closure of the orbit under translation of any such configuration supports exactly one ergodic translation-invariant measure on Ω, say μ, which is uniquely specified by the frequencies of all finite patterns. Such systems are called uniquely ergodic. The trivial example is the Ising antiferromagnet, where apart from the infinitely many defect ground state configurations there are two alternating ground state configurations but only one translation-invariant ground state measure which is just their average. Let us recall that a sequence of configurations converges if for any finite subset of the lattice all but a finite number of configurations are the same when restricted to this subset. Ergodicity of the measure means that for any local observable f the integral $\int f d\mu \equiv \mu(f)$, i.e., the average of the observable, is equal to $\lim_{n\to\infty} 1/N \sum_{i=1}^{N} f(T(x))$, where T is a shift operator, for μ-almost all x. For a uniquely ergodic measure the limit is uniform with respect to all choices of x. A uniquely ergodic measure can be a zero temperature limit of a low temperature Gibbs state (an infinite-volume grand canonical probability distribution) for an appropriate interaction. On the other extreme there is the example of the product (or Bernoulli) measure obtained by taking independently every spin to be $+$ or $-$ with probability

1/2 at every site. This translation-invariant measure corresponds to taking infinite temperature. With respect to this measure the frequency of plus spins equals 1/2 but has unbounded fluctuations, as large regions of pluses (and in fact any finite configuration) occur with finite (though small) density. In a typical "random" sequence these large fluctuations will certainly occur at various positions hence the closure of its orbit is the whole configuration space Ω.

If the system is uniquely ergodic (a property which is generic for ground states [14]) the support of its unique ergodic measure contains one minimal set, i.e., a non-empty, closed, translation-invariant subset of Ω which does not contain any proper subsets with this property. Physically, this means that all configurations containing defects have been eliminated. An example of such a configuration is a ground state configuration of the one-dimensional Ising antiferromagnet with spins being $\sigma_n = (-1)^n$ for $n \geq 0$ and $\sigma_n = (-1)^{n+1}$ for $n < 0$. The defect here is between -1 and 0. The property of minimality has been advocated by Aubry [6] under the name "weak periodicity" and has been discussed in [15] under the name "recurrence". Minimality means also that any pattern in a sequence appears again within a bounded distance. However, as asserted by Radin [16] in this connection, this property of minimality or even unique ergodicity does not imply anything in the sense of long-range order either in the sense of mixing properties (asymptotic independence at large distances) or spectral properties, as the Jewett-Krieger theorem tells us that any ergodic measure (including highly disordered ones) is isomorphic to a uniquely ergodic one [17, 18, 19].

One possibility of describing positional long-range order is to consider spectral properties of the shift operator T (considered as a unitary operator on $L^2(\mu)$). $L^2(\mu)$ is the space of functions which are square-integrable with respect to μ and is spanned by sums and products of spin values at finite sets of lattice sites. We write $Tf(x) = f(T(x))$. If the support of μ is small, many of these products coincide, viewed as elements of $L^2(\mu)$. For example, in the one-dimensional Ising antiferromagnet $\sigma_0\sigma_{2k} = 1$ for both periodic ground state configurations and any k, hence $\sigma_0\sigma_{2k}$ and 1 are in the same equivalence class. In fact, $L^2(\mu)$ is two-dimensional here and is spanned by σ_0 and 1. The simplest case of a one-dimensional lattice has been widely studied in ergodic theory [20]. We recall that for the study of the spectrum of the shift operator one considers generalized 2-point functions of the form $C_f(n) = \mu(fT^n f)$, where f can be any function in $L^2(\mu)$. By Bochner's theorem one can write $C_f(n) = \int_0^{2\pi} exp(2\pi i\lambda n)m_f(d\lambda)$, where m_f are measures on the interval $[0, 2\pi]$. If we consider higher dimensional lattices we get measures on $[0, 2\pi]^d$ and if we replace a lattice by a continuous space, measures on R^d. If for any choice of f, m_f has a pure point, singular continuous, or absolutely continuous part in its Lebesgue decomposition $m_f = m_{f\,pp} + m_{f\,sc} + m_{f\,ac}$, we say that the spectrum of the shift has a pure point, singular continuous, or absolutely continuous component. If the point spectrum consists solely of finitely many points one interprets this as a crystal; if one has a dense point spectrum one has a quasicrystal; and if there is also some (singular) continuous spectrum one might have a "turbulent" crystal [5]. When there is no long-range order only absolutely continuous spectrum is expected. However, there is always one discrete point in the spectrum at the origin. This is due to the fact that the constants are translation-invariant and thus are eigenfunctions of the shift operator.

In the second approach, which has been pursued by many people interested in the theory of quasicrystals, see for example [21, 22, 23, 15, 24, 25, 26, 27] one considers the Fourier spectrum (also called a structure factor or a correlation measure). This begins with the fact that diffraction patterns are given by the Fourier transform of the density of the diffracting matter. A structure factor $I(\overline{k})$ is proportional to the intensity of a spot $\overline{k}$ in the diffraction pattern and for one-dimensional systems $I(k) = |\sum_{n=-N}^{N} f(n) exp(2\pi ikn)|^2$, where $f(n)$ is an atomic scattering factor of an atom at a position n. For uniquely ergodic systems it can be shown [20] that $dkI(k)/(2N+1)$ converges weakly, as $N \to \infty$, to the so called correlation measure which is a spectral measure m_f of the shift operator corresponding to a special choice of $f = f(0)$ which evaluates sequences of atomic scattering factors at the origin (if we have only one type of atoms, $f(n)$ measures the presence of an atom at a position n and can be written $f(n) = \frac{1}{2}(\sigma_n + 1)$). m_f is then the Fourier transform of the 2-point function $\mu(f(0)f(n))$: $\mu(f(0)f(n)) = \int_0^{2\pi} exp(2\pi ikn)m_f(dk)$, where μ is the unique ergodic measure supported by the closure of the orbit of a configuration of diffracting matter. m_f is a measure on the interval $[0, 2\pi]$ and it will have a (quasi)crystalline character if it consists solely of (in)finitely many points in the pure point part of its Lebesgue decomposition.

We first point out that these two notions are not equivalent. We will illustrate this point on the example of the Thue-Morse system. Moreover, we observe that the first characterization has the advantage of allowing us to recognize molecular structures.

To define the Thue-Morse system we start by taking a sequence of all + spins ($\sigma_i = \pm 1, i \in Z$). At the first step we flip every second spin. At the nth step we flip all blocks of 2^{n-1} spins within the previous $n-1st$ configuration from the site $(2k+1)2^{n-1}+1$ to $(2k+2)2^{n-1}$ for every k.

A cluster point of this sequence of periodic configurations of period 2^n is a nonperiodic sequence called a Thue-Morse sequence. Its first 32 elements are

$$+--+-++--++-+--+-++-+--++--+-++-.$$

The closure of its orbit under translation supports exactly one ergodic translation-invariant measure μ_{TM} [28].

The Thue-Morse measure μ_{TM} was shown to be a unique ground state for arbitrarily fast decaying 4-body interactions [29]. Some aspects of its non-conventional long-range order were shown to persist at finite temperatures for some slowly decaying but still summable 4-body interactions [30]. In fact, a Thue-Morse sequence has been experimentally realized [31, 32] and the spectrum was observed to be singular continuous.

If one considers the Fourier transform of $C(n) = \mu_{TM}(\sigma_0\sigma_n)$ it is known to be singular continuous [34, 35, 28, 20]. Thus looking at the Fourier spectrum there would not be any quasicrystalline structure. However, if one considers the shift operator acting on the whole space $L^2(\mu_{TM})$ we find a richer behavior [28]. In particular, we have: $L^2(\mu_{TM}) = L^{2,odd}(\mu_{TM}) \oplus L^{2,even}(\mu_{TM})$, where the $L^{2,odd/even}(\mu_{TM})$ spaces are spanned by odd/even functions with respect to the spin flip operator $\sigma_i \rightarrow -\sigma_i$. T acting on $L^{2,odd}(\mu_{TM})$ has a singular continuous spectrum, while acting on $L^{2,even}(\mu_{TM})$ it has a dense point spectrum. Thus, if we look at correlation functions $C_f(n) = \mu_{TM}(fT^n f)$ with f even, for example $f = \sigma_0\sigma_1$, or more generally $f = \sigma^X$, $|X|$ even, its Fourier transform consists only of points $(\delta - peaks)$. The special choice $f = \sigma_0$ gives a singular continuous spectrum because of the oddness of σ_0. If we consider an X to be the shape of a molecule, these molecules can have

$\delta - peaks$ and can form more ordered structures than the underlying atoms. To be more precise: if ω is any Thue-Morse sequence then $s_A^n = T^n \sigma^A(\omega)$ for $|A|$ even is a q-periodic sequence according to the following definition [33]: A configuration of particles is q-periodic if, when a certain fraction of them is ignored, the rest of the configuration is periodic; the smaller the fraction the larger the period. By construction a Thue-Morse sequence is a sequence of blocks of spins M_k of length 2^m such that $M_k = \pm M(m)$ for some fixed block configuration $M(m)$ and this holds for all $m \geq 0$. Now, if one ignores lattice sites at the boundaries between consecutive $M_k' s$ (the density of which is $(diam(A) - 1)/2^m$) then s_A^n is a part of a periodic configuration of period 2^m, which shows that s_A^n is q-periodic.

As an example, if one places a delta function weight midway between each pair of equal adjacent spins and takes the Fourier transform of this object, it will consists entirely of delta peaks, whereas, if a delta function is placed midway between each pair of + neighboring spins, the Fourier transform will have both delta peaks and a continuous part, and finally, if delta functions are placed on the + spins and omitted from the − spins, the Fourier transform will have no delta peaks except the one at the origin. If a "molecule" is a pair of two neighboring equal spins the molecular spectrum is quasiperiodic, as $\delta_{\sigma_0 \sigma_1}$ is even, while a "molecule" consisting of two + neighbors gives rise to a mixed spectrum because $f = 1/4(\sigma_0 + 1)(\sigma_1 + 1) = 1/4(\sigma_0\sigma_1 + 1 + \sigma_0 + \sigma_1)$

has both an even and an odd part. Finally, placing delta functions just on the + spins (the atoms) corresponds to taking $f = \frac{1}{2}(\sigma_0 + 1)$ which is a sum of an odd function and a constant producing only a $\delta - peak$ at the origin.

For an example of a 2-dimensional system which has no $\delta - peaks$, at either atomic or molecular level, see [16] following [36]. This is in fact a unique ground state for a classical lattice gas model with a nearest neighbor interaction.

As for positive temperatures, there exists a 3-dimensional finite range ferromagnetic model due to Slawny [37] of a mixing Gibbs state, thus $\mu(fT^n f) \to (\mu(f))^2$ for all f, which has long-range order in the sense that it is not an extremal Gibbs state. It is not 3-mixing, that it is to say $\mu(fT^{n_1}fT^{n_2}f) \not\to (\mu(f))^3$ for some f and $|n_1|, |n_2|$ growing such that also $|n_1 - n_2| \to \infty$. This is an example of Ruelle's [5] proposed definition of a "turbulent" crystal, a system in which the decomposition into extremal Gibbs states is strictly finer than the almost periodic decomposition connected with the discrete part of the spectrum of the shift operator.

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