



WHAT IS . . .

a Quasicrystal?

Marjorie Senechal

The long answer is: no one is sure. But the short answer is straightforward: a quasicrystal is a crystal with forbidden symmetry. Forbidden, that is, by “The Crystallographic Restriction”, a theorem that confines the rotational symmetries of translation lattices in two- and three-dimensional Euclidean space to orders 2, 3, 4, and 6. This bedrock of theoretical solid-state science—the impossibility of five-fold symmetry in crystals can be traced, in the mineralogical literature, back to 1801—crumbled in 1984 when Dany Shechtman, a materials scientist working at what is now the National Institute of Standards and Technology, synthesized aluminium-manganese crystals with icosahedral symmetry. The term “quasicrystal”, hastily coined to label such theretofore unthinkable objects, suggests the confusions that Shechtman’s discovery sowed. What’s “quasi” about them? Are they sort-of-but-not-quite crystals? Solids with some sort of quasiperiodic structures? For that matter, what is a crystal?

Since the discovery of x-ray diffraction in 1912, a crystal’s identifying signature has been sharp bright spots in its diffraction pattern; that’s how Shechtman knew his were special. If it looks like a duck and quacks like a duck, it’s a duck: charged in 1992 with formulating a suitably inclusive definition, the International Union for Crystallography’s newly-formed Commission on Aperiodic Crystals decreed a *crystal* to be “any solid having an essentially discrete diffraction diagram.” In the special case that “three dimensional lattice periodicity can be considered to be absent,” the crystal is *aperiodic* (<http://www.iucr.org/iucr-top/iucr/cac.html>).

I was a member of the commission when this definition was hammered out, and I argued strongly in favor of it. It wasn’t a cop-out; it was designed to stimulate research. Which atomic structures or, more abstractly, point sets, have essentially discrete diffraction

diagrams? The set of vertices of a Penrose tiling does—that was known before Shechtman’s discovery. But what other objects do, and how can we tell? The question was wide open at that time, and I thought it unwise to replace one inadequate definition (the lattice) with another. That the commission still retains this definition today suggests the difficulty of the question we deliberately but implicitly posed. By now a great many kinds of aperiodic crystals have been grown in laboratories around the world; most of them are metals, alloys of two or three kinds of atoms—binary or ternary metallic phases. None of their structures has been “solved”. (For a survey of current research on real aperiodic crystals see, for example, the website of the international conference ICQ9, <http://www.icq9.ameslab.gov/index.html/>.)

And what have we learned about point set crystals? Let Λ be a discrete, countably infinite point set in R^n and μ_Λ the tempered distribution $\sum_{x \in \Lambda} \delta_x$. μ_Λ is a *Dirac comb*, N. G. de Bruijn’s apt name for any weighted sum of Dirac deltas $\sum_{w \in \Lambda'} c(w) \delta_w$, where Λ' is discrete. Whether Λ “diffracts” depends on the relative, not absolute, positions of its points, so we are less interested in Λ itself than in the interpoint differences $x - y \in \Lambda - \Lambda$ and in the convolution (autocorrelation) measure γ_Λ , a Dirac comb $\sum_{y \in \Lambda - \Lambda} c(y) \delta_y$ with nonnegative weights. The diffraction measure of Λ is the Fourier transform $\hat{\gamma}_\Lambda$. In general, $\hat{\gamma}_\Lambda$ is a sum of a Dirac comb $\hat{\gamma}_{discrete}$ and continuous components. The *diffraction diagram* of Λ is a plot of $\hat{\gamma}_\Lambda$; it is “essentially discrete” if $\hat{\gamma}_{discrete}$ is nontrivial, that is, if it has relatively dense support. (A set is relatively dense in R^n if its intersection with every ball of some fixed, sufficiently large, radius is nonempty.) If $\hat{\gamma}_\Lambda = \hat{\gamma}_{discrete}$, then Λ is a *pure point* crystal. A lattice L is the simplest pure point crystal: $L - L = L$ and, by the Poisson summation formula, $\hat{\gamma}_L = \hat{\gamma}_{discrete} = \sum_{x \in L^*} \delta_x$, where L^* is the lattice dual to L , $\{y \in R^n, \exp 2\pi i y \cdot x = 1, \forall x \in L\}$.

So far, so good, but we want to characterize crystals by local (geometrical) criteria, and this definition neither supplies nor implies them. In fact, some crystals are counterintuitive. We expect a point set crystal Λ to be, if not a lattice, then relatively dense. For

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The author would like to thank Robert Moody and the referees for helpful comments on earlier drafts.

example, the set of “visible points” (points with relatively prime coordinates) in the plane, which has “holes” of arbitrarily large radii, should not be a crystal. But it is, and pure point no less (note that $\Lambda - \Lambda$ is the integer lattice). On the other hand, some point sets that “ought” to be crystals are not. Consider the famous pinwheel tiling ([1], [3]) with congruent right triangles tiles. Like the Penrose tilings and many other aperiodic tilings (see the online “encyclopedia”, <http://saturn.math.uni-bielefeld.de>), the pinwheel tiling is a substitution tiling, generated by a straightforward two-step process, decomposition and inflation. But, unlike tile edges in a Penrose tiling, the triangles’ edges are aligned in a countable infinity of directions, so $\Lambda - \Lambda$ is not discrete.

Some large classes of aperiodic crystals do conform to our intuitive notions. Loosely speaking, point sets derived from lattices inherit a crystalline structure in modified form. If, like the set of visible points, Λ is a subset of a lattice L , then so is $\Lambda - \Lambda$ and $\hat{\gamma}_\Lambda$ is a weighted sum of deltas at points of L^* . We can construct a wide class of crystals Λ , the Penrose tiling vertices among them, from higher-dimensional lattices by the powerful and versatile cut-and-project method. Every cut-and-project set Λ is discrete, relatively dense, and a pure point crystal.

Cut-and-project sets are a special case of a large and general family of point sets called Meyer sets, which top a hierarchy of order types. Λ is a *Delone set* if it is uniformly discrete and relatively dense in R^n . A Delone set is of *finite type* if it has a finite number (finite atlas) of local patterns of every radius, up to translation. (Equivalently, Λ is of finite type if the difference set $\Lambda - \Lambda$ of interpoint vectors is closed and discrete.) A *Meyer set* Λ is a set of finite type where $\Lambda - \Lambda$ is Delone. Meyer sets are always crystals, though not necessarily pure point. They can be characterized in many ways; here is another: Meyer sets are almost-lattices. That is, Λ is a Meyer set if and only if, for every $\epsilon > 0$, its ϵ -dual $\Lambda^\epsilon := \{y \in R^n, |\exp 2\pi i y x - 1| < \epsilon, \forall x \in L\}$ is relatively dense.

But shouldn’t repetitivity (“quasiperiodicity”) be *the* local criterion we are looking for? I expect, when all is said and done, it will be, but its role is not yet clear. A Delone set Λ of finite type is *repetitive* if, for every $T > 0$, all local patterns of radius T are relatively dense [2]. The rate of growth, with increasing T , of the size of Λ ’s atlas of local patterns is a useful measure of its complexity. For example, if the growth function is bounded, then Λ is a lattice. One expects the diffraction condition to be characterized by some big-O condition on T . However, repetitivity lies somewhat outside the hierarchy sketched above: all repetitive Delone sets are of finite type, but not all repetitive sets are Meyer, and not all Meyer sets are repetitive. If a

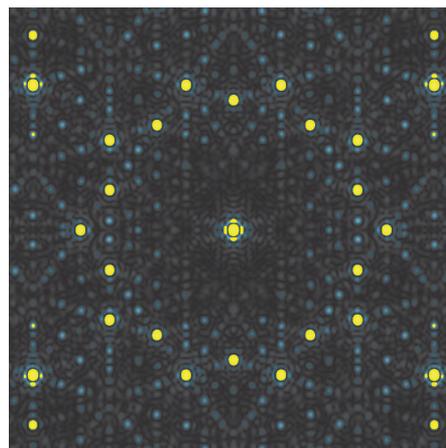
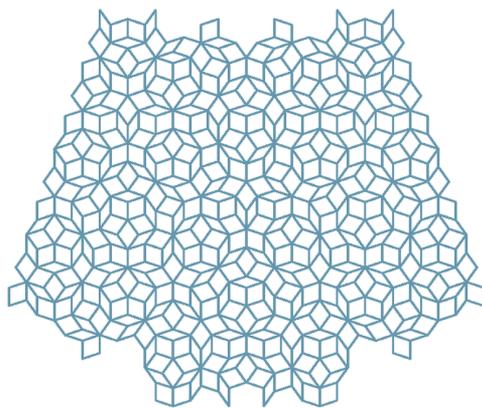


Figure 1. (a). A patch of a Penrose tiling. (b). The diffraction diagram of the vertex set V of (a) is essentially discrete; thus F is an aperiodic crystal, according to the new definition.

repetitive set is Meyer, then it’s a crystal since all Meyer sets are. But if it’s a crystal, is it Meyer?

Meanwhile the burgeoning mathematical field of long-range aperiodic order and the experimental study of real aperiodic crystals are symbiotic and mutually stimulating. Their cross-fertilization has been more metaphorical than practical, but no less valuable for that. Penrose tilings, the *Drosophila* of aperiodic order, don’t tell us what the structures of real aperiodic crystals are, but they do tell us what aperiodic order can look like. (Still, we are missing something. For suitable choices of lattice, dimension, and other parameters, we get cut-and-project sets with (diffraction) rotational symmetry of any finite order. Yet the symmetries of real aperiodic crystals found so far are only pentagonal, decagonal, dodecagonal, and icosahedral. Evidently, the real crystallographic restriction is yet to be discovered.)

Reciprocally, structure studies of real crystals suggest that coverings by clusters may be more useful models than packings and tilings. The Penrose tilings’ nonlocal growth (despite local matching rules) raises—but does not answer—questions about how crystals really grow. And recent experiments suggest that real crystal growth may be partly nonlocal too [4].

Stay tuned.

References

- [1] M. SENECHAL, *Quasicrystals and Geometry*, Cambridge Univ. Press, 1995.
- [2] J. C. LAGARIAS, Mathematical quasicrystals and the problem of diffraction, *Directions in Mathematical Quasicrystals*, M. Baake and R. Moody, eds., CRM Monograph series, Vol. 13, AMS, Providence RI, 2000, 61–93.
- [3] R. V. MOODY, Long-range order and diffraction, in *Proceedings of a Conference on Groups and Lie Algebras*, Ken-Ichi Shinoda, Sophia Kokyuroku, eds., in Mathematics 46, 2006. Available at <http://www.math.ualberta.ca/~rvmoody/rvm/rvmlpapers.html>.
- [4] C. DAY, Experiments vindicate a 50-year old explanation of how liquid metals resist solidification, *Physics Today* 7 (2003), 24–26; online <http://www.a.p.org/pt/vol-56/iss-7/p24.html>.