59. Ron Lifshitz

Senior Lecturer in Physics,
School of Physics & Astronomy,
Raymond and Beverly Sackler Faculty
of Exact Sciences, Tel Aviv University,
Tel Aviv 69978, Israel
ronlif@post.tau.ac.il
http://www.tau.ac.il/~ronlif/

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The Rebirth of Crystallography

Exactly 20 years have passed since the surprising discovery of the first quasicrystal by Shechtman [1]. Centuries of prior research in crystallography had led to the development of a beautiful and well-established science of crystals [2]. Modern crystallography was born in the 17th century thanks to the brilliant idea – attributed to such great scientists as Kepler and Hooke–that the observed properties of crystals were the result of internal order of "atomic" units. Unfortunately, by the 19th century, when Haüy began formulating the mathematical theory of crystallography, Kepler's insightful drawings of aperiodic tilings with decagonal symmetry were long forgotten [3, 4]. It became universally accepted, though never proven, that the internal order of crystals was achieved through a *periodic* filling of space. Crystallography treated "order" and "periodicity" synonymously, both serving equally to define the notion of a "crystal". With that came the so-called "crystallographic restriction", stating that crystals cannot have certain forbidden symmetries, such as 5-fold rotations. The periodic nature of crystals was "confirmed" with the discovery of x-ray crystallography and numerous other experimental techniques throughout the 20th century. The periodicity of crystals became the underlying paradigm not only for crystallography itself, but also for other disciplines such as materials science and solid state physics, whose most basic tools, like the Brillouin zone, relied on periodicity. By 1982 crystallography was a mature science, standing firmly on its theoretical paradigms, and offering sophisticated experimental tools for the structural study of materials. Shechtman's discovery of a crystal with 5-fold symmetry shook the science of crystals by its foundations. Crystallography was reborn.

On the occasion of the 10th anniversary of its announcement, Cahn [5] described the discovery of quasicrystals as a Kuhnian scientific revolution [6]. In my view, crystallography is now in the midst of the most exciting stage of this revolution. The old established paradigms, most importantly that of the periodicity of crystals, are being overthrown. The initial skepticism of the scientific community is being replaced by a growing acceptance. New notions and paradigms are being tested and carefully adopted. New theories of quasicrystals, aperiodic tilings, and symmetry are being developed. Experimental techniques are undergoing fundamental modifications to encompass aperiodic crystals. All this intense activity is being pursued by hundreds of scientists worldwide, ranging from pure mathematicians and crystallographers to physicists, chemists, and materials scientists. Although much progress has been achieved there is still more to be done. I believe that there are many years of exciting research still ahead before crystallography becomes, once again, a mature science awaiting its next revolution.

I would like to illustrate the difficulties, encountered in the current paradigm-building phase we are in, with three unresolved issues related to my own personal research in crystallography.

When it became clear that "periodicity" and "order" were not synonymous, a decision had to be made as to which would define the term "crystal". The International Union of Crystallography [7], through its Commission on Aperiodic Crystals, decided on the latter but was uncertain about the proper way to define "order". Clearly, periodicity was one way of achieving order, quasiperiodicity as in Penrose-like tilings was another, but can we be certain that there are no other ways that we have not yet discovered? The Commission found a clever solution to this dilemma by redefining "crystal" to mean "any solid having an essentially discrete diffraction diagram." The definition was shifted from a microscopic description of the crystal to a property of the data collected in a diffraction experiment. I am happy with this definition even though it still draws much criticism. It is consistent with the notion of long-range order, used in physics, whereby the transition from a disordered liquid to an ordered solid is indicated by the appearance of an "order parameter" – Bragg peaks in the diffraction diagram at non-zero wave vectors. It is sufficiently vague to serve as a temporary working definition until better understanding is obtained through additional research. In summary, crystals are ordered but not necessarily periodic. "Periodic crystals" form a well-understood subset, all the rest, commonly referred to as "aperiodic crystals", require further study.

Certain classes of aperiodic crystals were known long before Shechtman's discovery. These were the so-called "incommensurately-modulated crystals" and "incommensurate composite crystals" (or "intergrowth compounds"). These special quasiperiodic crystals did not pose any serious challenge to the periodicity paradigm because they could be viewed as periodic structures that had been slightly modified. Order was still obtained through periodicity – the paradigm remained intact. Shechtman's quasicrystal shattered the old paradigm because it violated the crystallographic restriction. It was clearly not a quasiperiodic modification of a periodic crystal, but rather a crystal which was intrinsically quasiperiodic. The observation of a forbidden symmetry in a crystal was so pivotal in starting this scientific revolution that it became the defining property of quasicrystals [8–10], even though there was no real reason to require that quasicrystals must possess forbidden symmetries. The crystallographic restriction was replaced by a "quasicrystallographic restriction". I have been arguing against this restriction on the definition of quasicrystals [11], and have been met with a surprising resistance, mainly from those who had been studying incommensurate crystals prior to the discovery of quasicrystals. It seems that there is yet another subtle paradigm in need of being overthrown, namely, that if a quasiperiodic crystal happens to have one of the 32 point groups, compatible with periodicity, it is necessarily a quasiperiodic modification of an underlying periodic structure. This is clearly not the case [12].

We have redefined the notion of a "crystal". Is it possible to avoid redefining the notion of "symmetry"? It turns out that quasicrystals, which clearly do not remain invariant under any translations, do not, in general, remain invariant under rotations as well. When a quasicrystal is rotated by one of its symmetry rotations, the rotated image is "indistinguishable" from the original one [13]. This means that the two images contain the same spatial distribution of bounded structures of any size. The two are statistically the same, but not necessarily identical. By using indistinguishability as the new defining criterion for symmetry, one obtains a straightforward extension of the traditional notions of "point group" and "space group" to quasiperiodic crystals [13–15]. As simple as it is, this approach to treating the symmetry of quasicrystals has not been accepted by most crystallographers. Instead, most prefer to describe the symmetry by embedding the crystal in a higher-dimensional "superspace", where its periodicity is recovered [16, 17]. This allows one to retain the traditional notion of a symmetry operation as one which leaves the (high-dimensional) crystal invariant. Although in certain circumstances there are practical advantages for using the superspace approach, the quasicrystal is a real 3-dimensional physical object. As such, one should be able to describe its symmetry without complicated and unnecessary mathematical detours into high-dimensional spaces. Mermin [18] has made the interesting analogy between this state of affairs in crystallography and that of astronomy at the time when Copernicus advocated his heliocentric view of the solar system. The prevailing paradigm that symmetry always means invariance does not work for quasicrystals. Overthrowing this paradigm may be the hardest struggle yet.

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