Comment on "Direct Imaging of Local Chemical Disorder and Columnar Vacancies in Ideal Decagonal Al-Ni-Co Quasicrystals"

Recently, Yan *et al.* [1] claimed to have provided direct evidence for the presence of a macroscopic amount of structural vacancies in an AlNiCo quasicrystal (QC). We suggest that this could be a misleading statement, as the empty sites in question do not comply with the proper definition of a vacancy. In fact, the authors observe pairs of atomic columns that are too close to be fully occupied simultaneously. Of the two adjacent sites these columns define in a plane, only one can be occupied at a time, since there is simply not enough space available to accommodate two atoms. An unoccupied site of this type cannot be called a vacancy if its twin site is occupied. An analogous situation occurs within the elementary octagon of an eightfold tiling with fat and skinny rhombi. Inside the basic octagon of this tiling we always find three vertices of the tiling. If the vertices are atomic positions, then there are eight different ways to orient these atomic triads within the interior of the octagon: They can be generated one from another by phason moves (see, e.g., Fig. 1 in [2]). The three atoms can visit in total eight positions defining a smaller, *virtual* octagon inside the initial octagon. One could thus loosely say that these eight interior "sites" have an occupation of $3/8$, but this does not imply that there would be a $5/8$ vacancy concentration, as the five "remaining sites" cannot possibly be filled with atoms. In fact, there are no vacancies at all in this perfect octagonal tiling, and the situation is better described by saying that there are three real sites and five virtual sites. The latter are nothing other than phason sites.

There are also physical grounds to be wary of the tacit redefinition of the vacancy concept contained in [1], as it may lead to confusion and contradictions. Physical data reveal that there is not a large amount of structural vacancies in QC, as is, e.g., the case in B2 phases which harbor a lot of them.

For certain stoichiometries the archetypal B2 phase AlNi can house up to 12% of *structural* vacancies at room temperature. This figure can be obtained by a comparison of the lattice parameter and density data [3]. While in QC the presence of voids prohibits this method, Mössbauer spectroscopy is a valid alternative. The presence of a vacancy leads to a larger electric-field gradient (EFG). Large vacancy concentrations are evidenced by a strong quadrupole-shifted additional component [4]. Similar spectroscopic data for QC (e.g., within the AlCuFe system) do not suggest the presence of a large structural-vacancy concentration [5]. The fact that the EFG in AlCuFe decreases smoothly with T shows that there is motional narrowing, but no other site or phase appearing [5].

The absence of a large concentration of structural vacancies in QC is confirmed by the totally disparate outlook of the existence domains of B2 phases and QC. The B2 structure is very insensitive to atomic composition as indicated by a sizable stability range within the phase diagram [3]. This hallmark of the presence of a large number of structural vacancies is not found in QC: On the contrary, they are extremely sensitive to composition [6]. Variations of 1% can destabilize the QC structure.

Finally, we must warn the reader that the mere presence of available sites is not sufficient to explain the physics of phason hopping in QC [7]. The temperature dependence of the quasielastic signal that betrays the presence of phason jumps, cannot be explained by just saying that they are thermally activated: The width of the signal is almost constant with temperature, while its intensity follows an Arrhenius plot. If the only criterion for having jumps were to be the presence of target sites, then it would be the width that exhibits the Arrhenius behavior, and the intensity that remains basically constant with temperature. This has been discussed by and large in Ref. [8]. Of course, it remains true that Yan *et al.* have achieved an experimental *tour de force.*

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