

In search of multipolar order on the Penrose tiling

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We use Monte Carlo calculations to analyse multipolar ordering on the Penrose tiling, relevant for two-dimensional molecular adsorbates on quasicrystalline surfaces and for nanomagnetic arrays. Our initial investigations are restricted to multipolar rotors of rank one through four – described by spherical harmonics Y_{lm} with $l = 1, \dots, 4$ and restricted to $m = 0$ – positioned on the vertices of the rhombic Penrose tiling. At first sight, the ground states of odd-parity multipoles seem to exhibit long-range order, in agreement with previous investigations of dipolar systems. Yet, careful analysis performed here establishes that, despite earlier claims, long-range order is absent for all types of rotors, and only short-range order exists. Nevertheless, we show here that short-range order suffices to yield a superstructure in the form of the decagonal Hexagon–Boat–Star tiling. Our results should be taken as a warning for any future analysis of order in either real or simulated arrangements of multipoles on quasiperiodic templates.

Keywords: multipolar order; adsorption; magnetic order; magnetism; order; quasicrystals; surface physics

1. Introduction and motivation

The concept of multipole moments is one of the most prominent and ageless mathematical constructions in physics and chemistry. Several decades ago the determination of electric and magnetic multipoles of neutral and polarized molecules became a vivid domain of scientific research because of the central role of multipole tensors for studies of intermolecular forces [1], nonlinear optical phenomena [2], electrostatic potentials [3], various phenomena induced by intermolecular forces [4], collision effects in nuclear magnetic resonance spectroscopy [5], hyperfine interactions [6], theoretical prediction of the geometries of van der Waals molecules [7] and electron scattering [8]. These extensive studies have demonstrated that many molecules possess sufficiently strong electric multipole moments. Among them are polar molecules with asymmetric charge distribution like HF, H₂O, FCl, HCCl₂, etc. having a permanent dipole moment; neutral Ne, Ar, Kr, Xe, O₂, F₂, D₂, CO₂, etc. possessing quadrupole moments; polyatomic SiF₄, B₄Cl₄, giant

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Keplerate molecule Fe_{30} and CF_4 having strong octupolar and $(\text{HSi})_8\text{O}_{12}$, $(\text{CH}_3\text{Si})_8\text{O}_{12}$ hexadecapolar contributions. Many organic substances possess multipolar moments as well. These are quadrupoles like benzene, 3, 4, 9, 10-perylenetetracarboxylicdianhydride (better known as PTCDA), cyanogen ($\text{N}\equiv\text{C}-\text{C}\equiv\text{N}$), 1,1-dichloroethene, *cis*-1,2-dichloroethene; octupoles like methane or cyanogen and more complicated complexes having higher order contributions [9,10]. Many of these molecules and molecular complexes can be adsorbed on solid surfaces. The arrays of adsorbates interact to a large extent via classical electrostatic multipolar interactions [10] as confirmed by Raman spectroscopy and nuclear magnetic resonance experiments. Often the multipolar interactions lead to complex ordering of the moments.

Quite independently, many new problems, that are not characteristic for bulk materials, arise at the nanoscale. One of the interesting aspects vividly discussed nowadays is the interparticle interaction in magnetic arrays. Magnetic properties of artificially structured and self-organized magnetic media belong to the central questions of nanomagnetism as they give access to new phenomena that can be used in technology [11–13]. Recently the importance of multipolar magnetostatic contributions for magnetization reversal in densely packed arrays of particles has been pointed out theoretically [14,15]. Here, the effect of the multipolar moments is two-fold. First, it influences the collective magnetic ordering in an array, and second, it changes the nucleation fields due to the stabilization of magnetization near the edges of neighbouring particles.

Hence, the knowledge of multipolar phase transitions and ground states is extremely important for a variety of applications as well as for fundamental understanding of the physics and the chemistry of solid state systems. With the recent ability to use the surfaces of real quasicrystals as templates upon which a variety of different particles can be adsorbed [16,17], it has become timely to study the ground states of multipoles on aperiodic substrates as well [18]. Yet, in contrast to the rather well-studied multipolar ground states on periodic lattices, the data for aperiodic tilings is quite limited [19–22], although there exists some group-theoretical analysis [23–28], describing the possible allowed symmetries of quasiperiodic multipolar arrangements, that may be used as a guide for our study.¹ The aim of this work is to initiate an extensive theoretical study of such order, beginning with a theoretical calculation of the ground states of multipolar rotors on the rhombic Penrose tiling.

2. Methods

In this study we investigate ground states of systems of multipoles, arranged on the vertices of the rhombic Penrose tiling, by means of Monte Carlo simulations. In order to calculate any order of interaction within reasonable effort, we introduce the Hamiltonian in spherical coordinates into the conventional MC scheme, and derive the stable low temperature configurations [36,37]. The Hamiltonian of the multipolar interaction reads

$$H = \frac{1}{4\pi\mu_0} \sum_{\substack{A \neq B \\ l_A l_B m_A m_B}} T_{l_A l_B m_A m_B}(\vec{R}_{AB}) Q_{l_A m_A}^A Q_{l_B m_B}^B \quad (1)$$

where $Q_{l_A m_A}^A$ and $Q_{l_B m_B}^B$ are the moments of multipoles A and B expressed in spherical harmonics, where l and m correspond to the standard two degrees of freedom on a sphere. The coupling coefficient $T_{l_A l_B m_A m_B}(\vec{R}_{AB})$ is the geometric interaction tensor

$$T_{l_A l_B m_A m_B}(\vec{R}_{AB}) = (-1)^{l_B} T_{l_A + l_B m_A + m_B}^*(\vec{R}_{AB}) \times \sqrt{\frac{(l_A + l_B - m_A - m_B)! (l_A + l_B + m_A + m_B)!}{(l_A - m_A)! (l_B - m_B)! (l_A + m_A)! (l_B + m_B)!}}, \quad (2)$$

where the dependence on the interparticle distance vector \vec{R}_{AB} , between multipoles on sites A and B , is given by the complex conjugate of the irregular normalized spherical harmonic function $I_{lm}(\vec{r}) = \sqrt{(4\pi/(2l+1))} (Y_{lm}(\theta, \phi)/r^{l+1})$.

For the sake of simplicity, the Monte Carlo simulations presented here are restricted to cylindrically symmetric multipole moments Q_{lm} , or *rotors*, with $l = 1, \dots, 4$ and $m = 0$, whereas generally m can take any value between $-l$ and l . These four multipolar rotors are depicted schematically in Figure 1, represented using corresponding charge distributions. The restriction to $m = 0$ is quite limiting and will be relaxed in future extensions of this study, nevertheless it is a good starting point providing interesting results. The moments are placed on the vertices of a finite patch of the two-dimensional rhombic Penrose tiling, using open boundary conditions. The patches are square or rectangular in shape, containing up to 1000 multipole moments. We also use circular patches to verify that our results are not affected by the shape of the sample. The simulations are performed with an algorithm especially designed for long-range systems: the local fields at each site are computed at the beginning of the simulation and are only updated when a rotation attempt is accepted [37]. To prevent artificial effects we do not use a cutoff in the evaluation of the multipolar coupling.

In contrast to MC schemes for usual magnetic systems, where only restricted rotations of the magnetic moment are often used [38], the rotational space is sampled continuously,

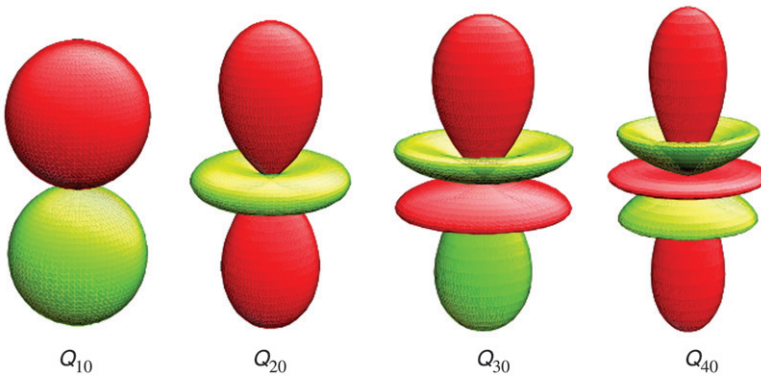


Figure 1. (Colour online) Cylindrically symmetric multipoles Q_{l0} with $l = 1, \dots, 4$, represented using an equipotential surface, with colour reflecting the sign of the charge on the internal side of the surface. Note that multipoles with even (odd) l are symmetric (antisymmetric) with respect to reflection about the equatorial plane. Thus, odd-parity multipole can be thought of as arrows \uparrow , while even-parity multipoles as double-headed arrows \updownarrow .

i.e. a moment can assume any new angle. This is especially important in complex multipolar arrays as these interactions might favour large angles between neighbouring spins. An extremely slow annealing procedure with up to 150 temperature steps is applied. To avoid metastable states we perform two different simulations of the same system simultaneously, starting at different seeds for the random number generator to ensure that the samples take different paths towards equilibrium. Only when both samples reach the same stable energy level is it taken for granted that the system has reached its equilibrium.

3. Ground states of classical multipolar rotors on the Penrose tiling

The symmetry of a charge distribution around a particle determines its non-zero multipole moments and whether moments of odd or even rank appear. The parity of the multipole moments has a strong impact on the ground state. We therefore consider the two cases separately below. As can be seen in Figure 1, once we restrict the value of m to be 0, the resulting multipoles with even l are symmetric with respect to the reflection about the equatorial plane, whereas those with odd parity are antisymmetric with respect to such a reflection. Consequently, odd-parity rotors can be represented geometrically as arrows, with light-coloured (yellowish-green online) ‘tails’ and dark-coloured (red online) ‘heads’, that like magnetic moments tend to align head-to-tail. On the other hand, even-parity rotors can be described as double-headed arrows with a repulsive interaction between the heads of neighbouring rotors. In the case of the Q_{20} rotors, the two heads are attracted exactly to the central oppositely-charged regions of nearby rotors. In the case of the Q_{40} rotors the attraction is not exactly to the centre, but rather to one of the two off-centre oppositely-charged regions. We establish below that these simple geometric observations suffice to explain all the calculated results that are presented here. Note that the charge flipping operation – a colour symmetry operation [23] that switches between red and yellow – is equivalent to the reflection about the equatorial plane, mentioned above, but only for the case of odd-parity rotors. In the case of even-parity rotors, charge flipping takes the rotors to oppositely charged counterparts that we do not consider in our simulations here. In the future it would be interesting to see what happens if one introduces charge flipping of individual moments as an additional MC step for the even-parity multipoles.

Experimental equivalents of the charge distributions having odd rank multipolar contributions include uniformly polarized magnetic and ferroelectric nanoparticles [15,39–42]. Generally, such particles may possess a mix of dipolar Q_{1m} , octupolar Q_{3m} , dotriacontapolar Q_{5m} and possibly even higher-order contributions. However, for certain geometries some of the multipole moments may become extinct. For example, a tetragonal prism with equal height, width, and length – which is therefore a cube – possesses strong dipolar but a zero octupole moment, while its strongly elongated or very flat counterparts have strong octupolar contributions. The dependence of the strength of multipole moments on the effective aspect ratio and shape of a particle can be found in [40,43]. To the experimental systems possessing multipoles of even order belong molecular adsorbates including H_2 , N_2 , CO on salts (e.g. boron nitride) or metal surfaces, organic PTCDA molecules on Ag, and methane on graphite.

3.1. Odd-parity multipole moments: dipoles and octupoles

In previous theoretical studies of dipolar ordering (multipole moments of rank one Q_{1m}) on the Penrose tiling, performed in Cartesian coordinates [20,44,45], a decagonal pattern with long-range order was proposed as the ground state. Here, after much further analysis, we find clear evidence for short-range order, with very interesting geometric properties, yet we see no evidence for the emergence of long-range multipolar order. The ground-state configurations of odd-parity multipoles are shown in Figure 2. At first sight these configurations seem to possess very nice long-range multipolar order, as one clearly sees an arrangement of decagonal rings of multipoles. In fact, as we note here for the first time, if one looks more carefully one can actually identify a perfect superstructure in the form of the decagonal Hexagon–Boat–Star (HBS) tiling [46]. Each decagonal ring is subdivided into a single boat and a pair of hexagonal tiles, and between the decagons one can see the star-shaped tiles. This aesthetic arrangement of the multipoles may lead one to the incorrect conclusion that there exists long-range multipolar order on the underlying Penrose tiling. Yet further analysis, as described below, shows that this arrangement stems from the short-range head-to-tail attraction of neighbouring multipoles and exhibits *no long-range order*. We would like to use this example as a warning for any future analysis of either real or simulated arrangements of multipoles on quasiperiodic templates. One has to be very careful in the analysis of order in such quasiperiodic structures and explicitly calculate their Fourier spectra, as it is harder to visually comprehend them in real space than their periodic counterparts.

Looking closely at the edges of the HBS tiles, we see that the HBS tiling is simply outlined by pairs and triplets of multipoles that are separated by the short diagonals of the thin (36°) rhombic tiles of the Penrose tiling. This separation, which is the shortest interparticle separation on the Penrose tiling, sets the largest energy scale in the system.

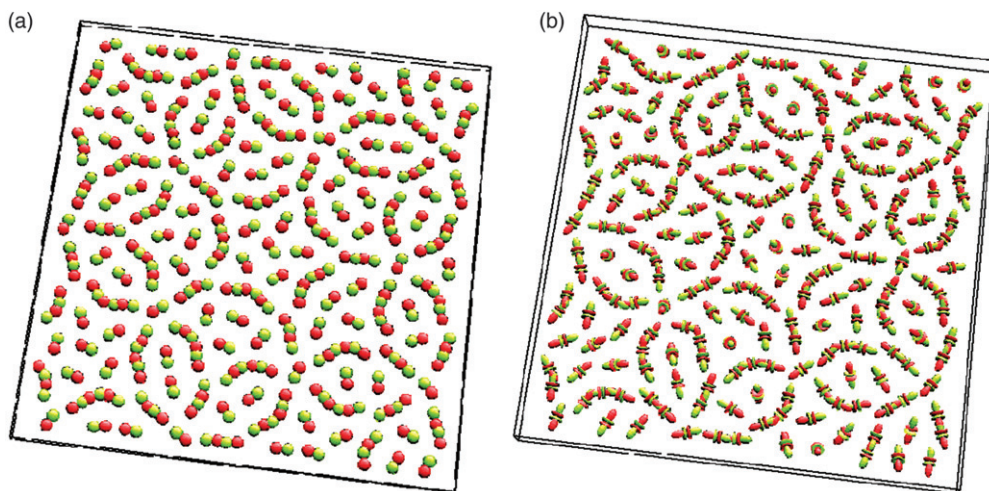


Figure 2. (Colour online) Ground-state configurations of odd-parity multipolar rotors. (a) dipole moments; (b) octupole moments. These configurations possess short-range order, owing to the strong head-to-tail attraction of neighbouring multipoles, which is sufficient to highlight the decagonal Hexagon–Boat–Star tiling.

As such, these pair and triplet chains are the first to order as the temperature is lowered. Because their positions are strictly inherited from the Penrose tiling, and their orientations dictated by the head-to-tail attraction described earlier, their ordering on the short scale suffices to outline the HBS tiling that one clearly observes. The existence of short-range order in the orientation of the multipoles can be verified quantitatively through a statistical analysis. The absolute orientation θ of the multipoles, projected onto the plane, is clearly peaked along the twenty directions ($n\pi/10$ for $n=1, \dots, 20$), dictated by the Penrose tiling, as shown in Figure 3(a). This is a direct outcome of the simple head-to-tail attraction of nearest neighbours. The dipolar histogram is more strongly peaked relative to the octupolar one owing to the fact that the octupolar ground state possesses, on average, a larger out-of-plane component. The frequency distribution of the angle ϕ between nearest neighbouring moments, shown in Figure 3(b), is also peaked at the characteristic angles inherited from the relative orientation of thin rhombic tiles on the Penrose tiling, and is significantly less-pronounced for the octupolar moments due to their substantial out-of-plane protrusion. We note that other than the fact that the octupolar arrangement contains a larger average out-of-plane component, and a less-perfect short-range order within the plane, the two cases are quite similar. The difference stems from the fact that there is some amount of attraction of the arrow heads to the central, oppositely-charged, regions of neighbouring octupoles (see Figure 1).

We note that in magnetic systems the strength of the multipolar interactions can be tuned by the shape of the particles, or their size relative to the interparticle separations. The octupolar contribution may become very large for $R_{AB} < s$, with s being the lateral size of a particle. The dipolar contributions are sizable for $R_{AB} < 5s$ to $10s$. For very small interparticle separations the decagonal structure might become disordered due to the octupolar contributions, while for very large separations disorder may appear because of the weakness of the dipolar coupling. This implies that there exists a critical separation R_{AB}^c for which the short-range ordering of odd-parity multipoles on the Penrose tiling is maximal. For typical particle shapes used in experiments [37] this critical distance is of order of $1s$ to $2s$.

The multipoles that lie within the HBS tiles are disordered, as can be verified by simple inspection. Nevertheless, one could still imagine a situation in which the multipoles that lie on the edges of the HBS tiles are long-range ordered while the internal multipoles are not. Yet upon further inspection one finds that multipoles that lie on the edges of the HBS tiles are disordered as well, as their direction changes randomly from one pair or triplet chain to the next. This disorder is a direct consequence of the frustration that arises whenever the

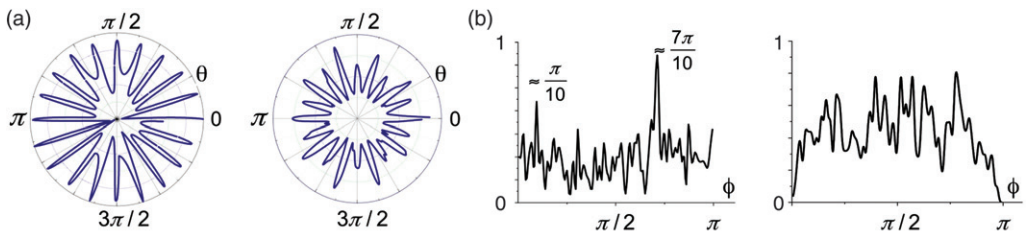


Figure 3. Angular distributions. (a) Absolute in-plane orientation angle θ of dipoles (left) and octupoles (right). (b) Relative angle ϕ between neighbouring dipoles (left) and octupoles (right).

ends of three such chains meet together. This can be seen, for example, at the 5 vertices of the central star tile in both configurations, shown in Figure 2. Our observation of the lack of long-range order is confirmed quantitatively by performing a Fourier analysis of the ground state configurations. Examples of such calculations for the octupolar case are shown in Figure 4. By examining the different components of the multipolar fields, as well as various functions of the components, we can say with certainty – despite all previous reports – that such order is lacking, as the calculated Fourier spectra show no additional Bragg peaks when compared to the Fourier spectrum of the tiling itself, shown in Figure 4(a). Thus, the only long-range order that is observed is in the positions of the multipoles, inherited from the Penrose tiling, and not from their relative orientation. To be sure, we have also calculated the Fourier spectra of randomly oriented configurations of multipole moments on the vertices of the Penrose tiling, shown for the octupolar case in Figure 4(b), created using a random number generator. The outcome strongly resembles those of the ground-state configurations.

A natural question to ask at this point is whether the lack of long-range order of our arrow-like objects on the Penrose tiling is a geometrical property of the tiling. If we disregard the physics, or the energetics, would it be possible to find a geometrical arrangement of ordered arrows on the Penrose tiling? The positive answer to this question was given by one of us many years ago [47], where two such configurations – in which the arrows are located at the tile centres, rather than their vertices – were demonstrated and analysed using the tools of colour [23] and magnetic [24,28] symmetry. If this is the case, then one should wonder whether our lack of order is a physical consequence of the particular type of interaction Hamiltonian (1) that we use. Would it be possible to arrange arrow-like objects on the Penrose tiling with sufficiently weak frustration, or possibly no frustration at all, and obtain an ordered ground state of some other Hamiltonian? It turns out that the answer to this question is also yes, if we just realize that we can map our system to one that was studied almost a decade ago by Cockayne and Widom [46]. In their system – a model of a decagonal Al–Cu–Co quasicrystal – there were no arrows *per se*, but one can identify abstract arrows in their model that are positioned on the edges of the Penrose tiles, representing the chemical ordering of pairs of Cu and Co atoms. Thus, if one imagines the yellow- and red-coloured circles of our dipole in Figure 1 (left) as being copper and cobalt atoms, one can obtain the desired ground state using the model of Cockayne and Widom. It remains to be seen whether an appropriate modification of our interaction Hamiltonian, can lead to an ordered ground state of multipoles.

3.2. *Even-parity multipole moments: quadrupoles and hexadecapoles*

Patches taken from the ground-state configurations of even-parity multipoles are shown in Figure 5. These configurations do not possess any long-range order, as confirmed by a similar Fourier analysis. Short-range order is clearly present, but it is not as easy to analyse and quantify as the case of odd-order multipoles where it shows up in the form of the HBS superstructure. Nevertheless, one can see that the quadrupole moments tend to align predominantly at 90° angles, as well as 72° , owing to the strong attraction of the edges of one quadrupole to the centre of its neighbouring quadrupole. In this manner the quadrupoles can form nicely-ordered local decagonal structures, like the one seen at the centre of Figure 5(a). The local arrangements of the hexadecapoles are even more complicated to

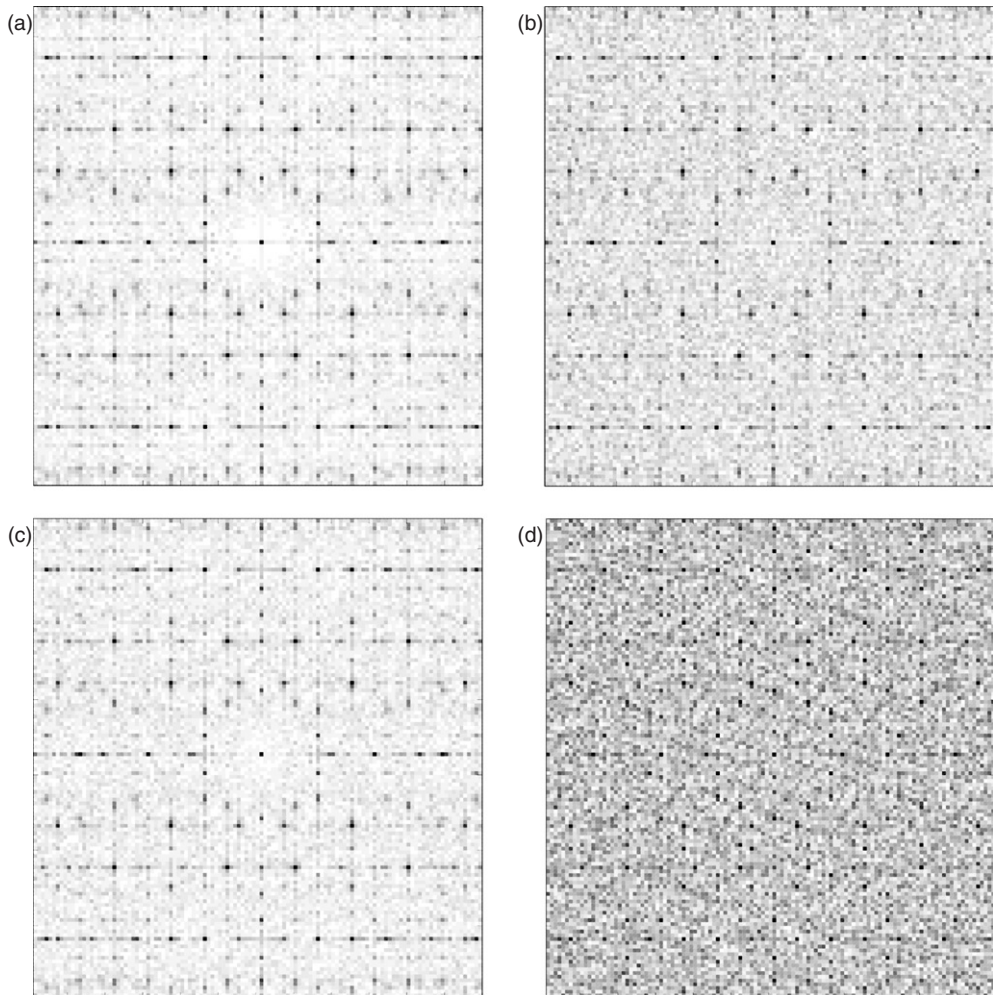


Figure 4. Representative Fourier spectra of octupolar arrangements on the vertices of the rhombic Penrose tiling, calculated using a numerical fast Fourier transform. (a) Fourier spectrum of the tiling itself, obtained by positioning point particles on the vertices of the tiling. The diffuse background is a numerical artifact. (b) Fourier spectrum of the function $S_x^2 + S_y^2$ for randomly oriented moments. Note that the contrast between the Bragg peaks and the background is weaker in this case because of the disorder in the octupolar orientations. (c) Fourier spectrum of the function $S_x^2 + S_y^2$ for the ground state shown in Figure 2(b). There is no appearance of additional Bragg peaks that are not already inherited from the tiling itself, indicating no octupolar long-range order. The contrast is slightly reduced, indicating a certain degree of disorder. (d) Fourier spectrum of the function S_z^2 for the ground state shown in Figure 2(b). No additional Bragg peaks appear here as well and the contrast is even weaker owing to the smaller and disordered out-of-plane components of the octupoles.

describe, with the edges of one attracting the opposite charges located slightly off the centres of its neighbours. Nevertheless, one can see nice triplet chains of hexadecapoles, and an overall nicely-ordered local pentagonal configuration like the one shown in Figure 5(b). As mentioned above, this order does not extend beyond the short range.

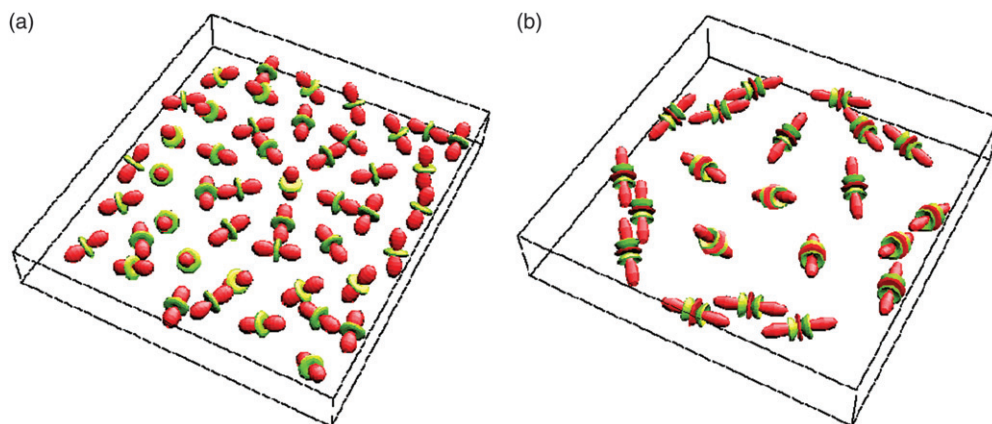


Figure 5. (Colour online) Ground-state configurations of even-parity multipolar rotors. (a) quadrupole moments; (b) hexadecapole moments. These configurations possess some degree of short-range order which is harder to establish than in the case of odd-parity multipoles of Figure 2. One clearly sees that the red-coloured edges are attracted to the central, or slightly off-centred, yellow regions of neighbouring multipoles. This causes the quadrupoles to align at exactly 90° , taking advantage of out-of-plane orientations. In the case of hexadecapoles the local arrangements are less trivial, although they clearly exhibit local symmetry.

4. Summary

In conclusion, we have studied the low-temperature stable multipolar ground states on the Penrose tiling by theoretical means. We have shown here that long-range order is absent in all cases we examined despite earlier claims about dipolar order. Nevertheless, short-range order exists owing to the strong interaction between particles of closest separation – those that are separated by the short diagonals of the thin Penrose tiles. In the case of odd-parity multipoles, we demonstrate here that this short-range order, combined with the underlying structure of the rhombic Penrose tiling, suffices to outline a superstructure in the form of the decagonal HBS tiling. The multipoles lack long-range order despite the appearance of the HBS superstructure, because as we show above the orientations of the moments on the edges of the HBS tiles are disordered due to three-body frustration.

Further investigations are clearly necessary in order to seek out other possibilities of long-range multipolar order on the Penrose tiling. These should relax the $m = 0$ restriction imposed here, allow for charge flipping of even multipoles, consider alternative forms of interparticle interaction, as well as explore other possibilities for the positions of particles on the quasiperiodic surface, such as those corresponding to the so-called ‘dark-stars’ on the 5-fold surfaces of real icosahedral quasicrystals [16–18,48].

Acknowledgements

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Note

1. Some related results exist for studies of quantum magnetic models on quasicrystals [29–34]. See also the discussion of magnetism in quasicrystals in this issue [35].

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