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# Interplay between magnetic and spatial order in quasicrystals

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The stable magnetisation configurations of antiferromagnets on quasiperiodic tilings are investigated theoretically. The exchange coupling is assumed to decrease exponentially with the distance between magnetic moments. It is demonstrated that the combination of geometric frustration and the quasiperiodic order of atoms leads to complicated noncollinear ground states. The structure can be divided into subtilings of different energies. The symmetry of the subtilings depends on the quasiperiodic order of magnetic moments. The subtilings are spatially ordered. However, the magnetic ordering of the subtilings in general does not correspond to their spatial arrangements. While subtilings of low energy are magnetically ordered, those of high energy can be completely disordered due to local magnetic frustration.

## 1.   Introduction

In contrast to the rather well-studied spin structure of antiferromagnets on periodic lattices, the antiferromagnetic ordering of quasicrystals is subject of ongoing scientific debate [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. Experimentally, it has been demonstrated that rare earth containing quasicrystals exhibit spin glass-like freezing at low temperatures [3, 5]. However, this freezing is different from that of conventional spin glasses. The observed dependence of the thermoremanent magnetisation on the magnetic field does not follow the spin-glass behaviour and the frequency shift of the freezing temperature lies between those of a canonical spin glass and of a superparamagnet [7]. Hence, the free energy landscape of a rare earth quasicrystal is different from both the highly degenerate distribution of energy barriers in spin glasses and the single global energy minimum in superparamagnets.

Although the atomic and electronic structure of rare earth quasicrystals is not completely understood, it has been postulated [7] that the low-temperature microstructure of such a magnet resembles geometrically frustrated but site-ordered magnetic systems and consists of weakly interacting magnetically ordered clusters. Another interesting approach is based on recent elastic neutron scattering experiments on a Zn-Mg-Ho icosahedral quasicrystal [6] revealing a very peculiar diffuse scattering pattern with icosahedral

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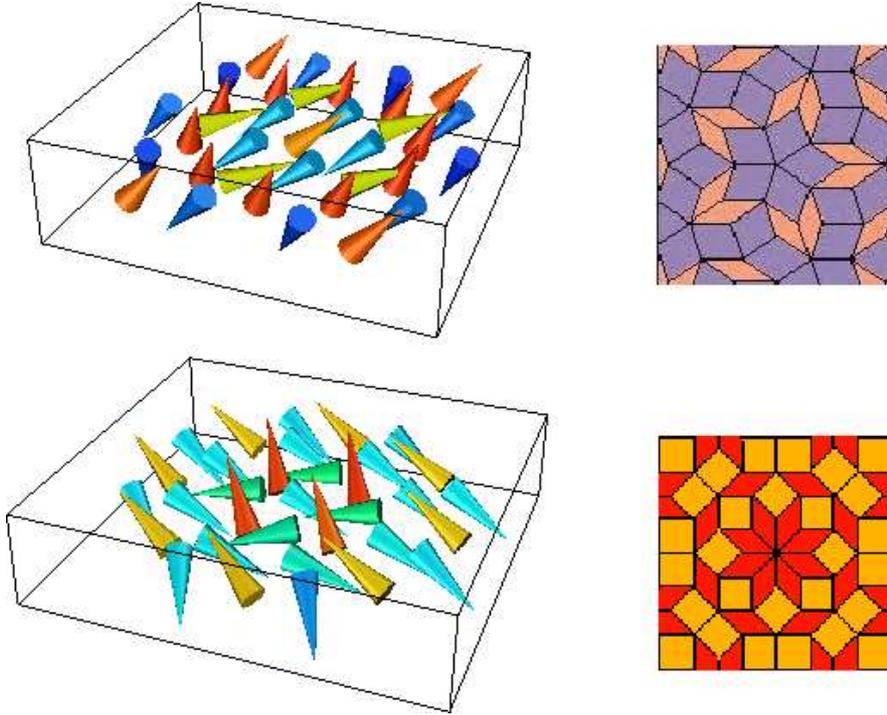‡Corresponding author. Email: u.g.grimm@open.ac.uk

Figure 1: Perspective view of a portion of a Monte-Carlo configuration on the Penrose tiling (top) and the octagonal tiling (bottom). Top views of the corresponding patches are shown on the right. The magnetic moments are represented as cones.

symmetry at temperatures below 6K. In contrast to reference [7], the authors interpret the diffraction pattern as that of several interpenetrating quasiperiodic sublattices, where all spins point in the same direction [10]. Recent theoretical studies of real-space magnetic configurations on the octagonal tiling [8, 10, 11] demonstrate that the energy landscape, in accordance with [7], is neither degenerate nor has a single global minimum. All spins can be divided into several quasiperiodic (in the 2D physical space) or periodic (in the corresponding 4D periodic hypercrystal) subtilings of different energy.

In the present investigation, we calculate the low-temperature stable antiferromagnetic configurations on several planar quasiperiodic tilings with tenfold symmetry. In most rare earth intermetallic compounds an oscillatory (RKKY - like) exchange interaction has been observed. To tackle this complicated problem first we concentrate on exponentially decreasing exchange coupling corresponding to a rapid-decaying limit of an oscillatory interaction. It will be demonstrated that the real-space magnetic structure is generally three-dimensional and noncollinear. In disagreement with [7], and in accordance with [6], the magnetic structure consists of several ordered interpenetrating quasilattices with characteristic wave vectors.

## 2. Simulations and results

We have investigated the magnetic ordering in an antiferromagnet on Penrose, Anti-Penrose, Tübingen triangle [12] and Tie-Navette [13] tilings by means of Monte-Carlo simulations. Two-dimensional films of classical, three-dimensional magnetic moments **S**
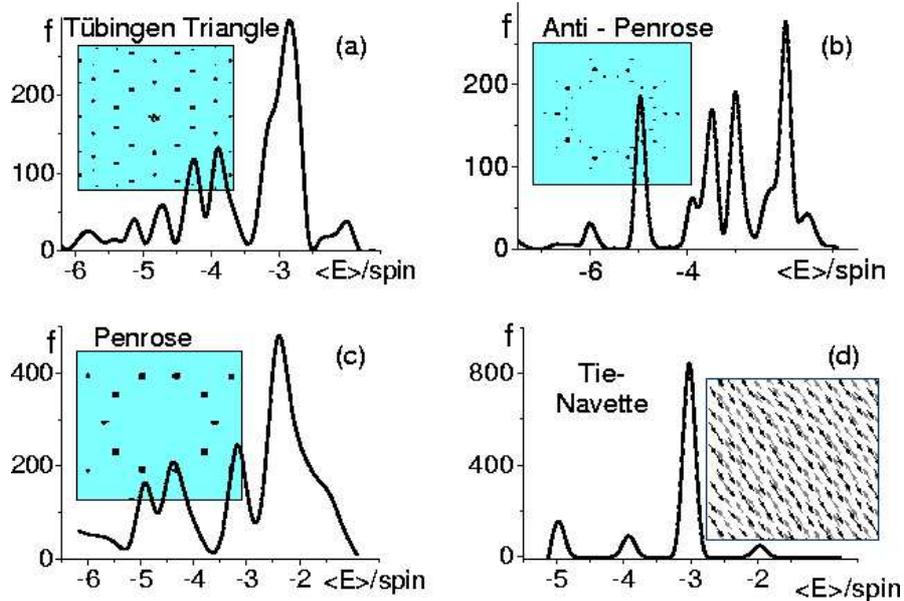
Figure 2: The frequency distribution of the energy per spin on the Tübingen triangle (a), Anti-Penrose (b), Penrose (c) and Tie-Navette (d) tilings for classical vector spins. A purely antiferromagnetic interaction $J$ at a temperature $kT = 0.01J$ is considered. The insets in (a)–(c) give the calculated Bragg scattering of the $S^y$ component of the magnetisation for subtilings composed of magnetic moments belonging to peaks with $-6 < \frac{\langle E \rangle}{spin} < -4$. The scale goes from -6 to 6 $k_{x,y}^{S_y}/\pi$. The inset in (d) shows a portion of the stable magnetic configuration on the Tie-Navette tiling as described in the text. Dark and light grey arrows denote antiparallel magnetic moments.

have been studied. The Hamiltonian of the problem is given by

$$H = J_{ij} \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - K_1 \sum_i (\mathbf{S}_i^z)^2 \qquad (1)$$

where $J_{ij}$ are the exchange coupling constants and $\langle i,j \rangle$ refers to pairs of spins. Two cases have been explored: $J_{ij} = 1$ for all $r_{ij} \leq 1$ (and $J_{ij} = 0$ for all $r_{ij} > 1$), and an exponential decrease of the exchange interaction with the distance between magnetic moments (which for practical purposes was cut off at distance $r_{ij} > 2$), where $r_{ij}$ denotes the distance between sites $i$ and $j$ (as compared to the edge length in the tiling, which are chosen to have length one). The samples are patches of square or rectangular shape, containing some 10 500 magnetic moments. We also used circular areas to check that our results are not affected by the shape of the sample. An extremely slow annealing procedure, with 50 temperature steps per Monte-Carlo run, has been applied. To see the time-dependent changes in a microstructure, we ran the simulation for several hundred thousand steps per temperature.

In previous theoretical studies [1, 2, 4] frustrated, two-dimensional structures have been proposed. In accordance with previous publications, we find that the ground state of a system with purely antiferromagnetic exchange interactions is locally frustrated. Under the local frustration $f$ we understand the normalised difference between an actual energy $E_i$ of a spin $i$ and a ground state energy $E_{id}$ of a relevant unfrustrated vertex
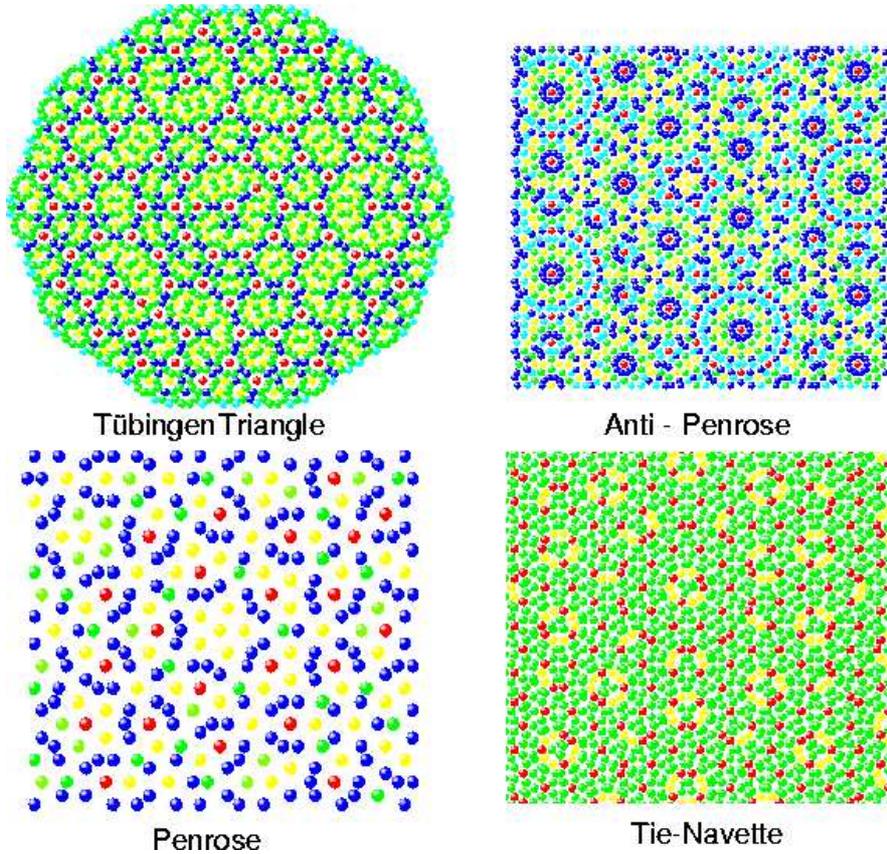
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Figure 3: Energy maps for classical vector spins on Tübingen triangle (a), Anti-Penrose (b), Penrose (c) and Tie-Navette (d) tilings. The circles give positions of magnetic moments. Different shades of grey denote different energies corresponding to the peaks in figure 2. Purely antiferromagnetic interaction with $J = 1$ for all $r_{ij} \leq 1$ at $kT = 0.01J$ is considered.

with all spins antiparallel to the spin $i$

$$f = \frac{|E_{id}| - |E_i|}{|E_{id}|}. \tag{2}$$

In contrast to the common folklore, the configurations are three-dimensional. Similar to the underlying atomic symmetry, the magnetic structure is quasiperiodic, i.e. it consists of identical units which do not have identical surroundings.

Three-dimensional representations of parts of the low-temperature quasiperiodic patterns observed for the Penrose and the octagonal tiling are shown in figure 1. The corresponding configurations represent the characteristic Penrose and Amman-Beenker 'stars', which are also shown in figure 1 for clarity. On the Penrose tiling, the 'star'-pattern can easily be recognised in the magnetic structure, because the moments belonging to the perimeter of enclosed 'stars' show perfectly antiparallel alignment. On the octagonal tiling, the situation is more complicated. The central magnetic moment is neither parallel nor antiparallel to the neighbouring magnetic moments. Its eight nearest neighbours have different sets of mutual angles. The moments forming the next ring have still another orientation with respect to their nearest neighbours. The noncollinear alignment of the neighbouring moments indicates that the system is geometrically frustrated, i.e.

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there is no possibility to align all neighbours in an antiparallel arrangement. Similar noncollinear antiferromagnetic configurations are formed in the Tübingen triangle and Anti-Penrose tilings. Within the examples of tilings considered here, the Tie-Navette tiling represents an exception. The magnetic structure observed for this tiling consists of two antiferromagnetically aligned quasiperiodic sublattices, as shown in figure 2(d). This means that every pair of nearest neighbouring moments can be aligned antiparallelly, i.e. the antiferromagnetic configuration is not frustrated.

We have calculated the stable low-temperature configurations and the frequency distribution of the exchange energy per atom $\langle E \rangle$ for the Tübingen triangle, Anti-Penrose, Penrose and Tie-Navette tilings. The calculations have been performed for an exponentially decreasing exchange coupling and for a short-range exchange coupling $J_{ij} = \text{const} = 1$ for all $r_{ij} \leq 1$. The analysis of the local energies reveals several characteristic energetic maxima in the frequency distributions shown in figure 2(a)–(d). The magnetic configurations and the number of the energy peaks for the same tiling are identical for both choices of exchange couplings ($J_{ij} \propto e^{-r_{ij}}$ and $J_{ij} = 1$ for $r_{ij} \leq 1$). For different tilings, the number and the width of the maxima are different. The simple existence of the peaks means that there exist different sorts of magnetic moments having well-defined relative orientations with respect to their nearest neighbours. These relative orientations depend on the tiling and *not* on the choice of the exchange couplings $J_{ij}$. For $J(r_{ij} \leq 1) = 1$, however, it can be seen directly from the energy distributions of figure 2, whether the magnetic ordering is collinear or noncollinear. If all nearest neighbours are collinear (parallel or antiparallel), then the exchange energy per spin should have integral values depending only on the number of the neighbouring moments. This is indeed the case for the Tie-Navette tiling; compare figure 2(d). For a noncollinear alignment of neighbouring magnetic moments, $\langle E \rangle$ should be non-integral as the cosines of the angles between the moments are no longer zero or unity. This happens for all other tilings we considered; compare figure 2(a)–(c). The average energy of noncollinear configurations is smaller than the energy of any collinear solution. Hence, the increase of the configurational entropy permits to minimise the average local frustration and the total energy of the system.

The spatial arrangements of the exchange energies of the magnetic moments are given in figure 3. Each shade of grey in figure 3 represents a certain energy range corresponding to one of the peaks in the spectra of figure 2. The magnetic moments form subtilings of different energies, which generally do not coincide with a tiling obtained by selecting a specific vertex type. The subtilings of low energy $\frac{\langle E \rangle}{spin} < -3$ are magnetically stable and ordered while those of higher energy $\frac{\langle E \rangle}{spin} > -3$ disordered. The disorder can be seen in the portion of the magnetic configuration shown at the bottom of figure 1. The two front moments belonging to the subtiling of a large energy have angles which deviate considerably from those of the other moments in the ring while the moments in the inner rings with lower energy have collinear orientations. With increasing temperature the magnetisation of subtilings of large energy is fluctuating while the magnetisation of low-energy subtilings is still stable. The spatial quasiperiodic ten-fold symmetry of the ordered subtilings can be seen from the calculated magnetic Bragg scattering given in the insets to figure 2. While the atomic ordering of the unstable subtilings can be seen in the Fourier space their magnetic reflexes are extinct because of disorder.

## 3. Summary

In conclusion, we demonstrate that vector spin system with antiferromagnetic coupling on different quasiperiodic tilings is locally frustrated. All spins can be divided into several quasiperiodic (in our two-dimensional physical space) or periodic (in the corresponding four-dimensional periodic hypercrystal) subtilings of different energy, which generally do not coincide with a specific vertex type. The vector spin system admits a three-dimensional noncollinear magnetic structure. The noncollinearity of the magnetic configuration permits to minimise the degree of frustration and the total energy of the system in comparison with the collinear case. The co-directional spins of every subtiling reveal quasiperiodic ordering with a wave vector which is specific for a given subtiling. The Tie-Navette tiling is not frustrated and admits collinear magnetic configurations. For the short-ranged exchange interaction, this arises as a consequence of the bipartiteness of the graph formed by connecting interacting pairs of spins; however, we observe that the antiferromagnetic order persists for the case of a long-range, exponentially decreasing exchange interaction.

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