Decagonal Quasiferromagnetic Microstructure on the Penrose Tiling

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The stable magnetization configurations of a ferromagnet on a quasiperiodic tiling have been derived theoretically. The magnetization configuration is investigated as a function of the ratio of the exchange to the dipolar energy. The exchange coupling is assumed to decrease exponentially with the distance between magnetic moments. It is demonstrated that for a weak exchange interaction the new structure, the quasiferromagnetic decagonal configuration, corresponds to the minimum of the free energy. The decagonal state represents a new class of frustrated systems where the degenerated ground state is aperiodic and consists of two parts: ordered decagon rings and disordered spin-glass-like phase inside the decagons.

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There is currently a broad interest in the understanding of the magnetism of ultrathin magnetic structures due to the wide variety of industrial applications [1]. The discovery of the rare-earth-based quasicrystals [2] offers the unique opportunity to study the magnetic behavior of localized magnetic moments in magnets with nonperiodic structure. The combination of the structural quasiperiodicity with magnetic properties of ultrathin films can lead to new physical phenomena. Hence, the understanding of the micromagnetic ordering in such objects is of high significance for the fundamental physics of magnetic materials as well as for technological applications.

The critical behavior of localized magnetic moments on quasiperiodic tilings has been investigated theoretically [3]. In those studies emphasis has been put on critical exponents and transition temperatures of Ising, Potts, and *XY* models. In the investigations only the short-range exchange interaction has been taken into account. The long-range dipolar forces were not considered. On the other hand, due to the long-range character, a relatively weak dipolar interaction can compete with the strong but short-range exchange coupling [4]. The competition can lead to a variety of magnetic configurations in two-dimensional films [4]. In quasiperiodic magnets the magnetic pattern will be different from that of periodic crystals and disordered media.

The quasicrystals can be structurally ranked between the periodic lattices and completely disordered media. In contrast to periodic crystals, in quasicrystals the number of nearest neighbors varies widely from one point to another as in disordered matter. The Penrose tiling [5], for example, has atoms with coordination number changing from 3 to 7. Hence, the energy per magnetic moment also varies. Unlike the disordered media, however, this variation exhibits a long-range orientational order, i.e., any finite section of a quasicrystal is reproduced within a certain distance. In particular, fivefold symmetry, forbidden in conventional crystallography, can be observed in the diffraction patterns. Thus, the magnetic ordering in quasicrystals should be different from the collinear magnetism of periodic crystals and from spin-glass-like behavior of the disordered media.

The dipolar system on a Penrose tiling is geometrically frustrated; i.e., magnetic moments are unable to find an orientation satisfying the interactions with all neighbors. The frustration in quasicrystals is different from that of periodic systems and that of disordered media. In highly ordered magnets the frustration is uniform, i.e., equal for all lattice points. In disordered materials the frustration is random. In quasicrystals the change in coordination number leads to spatial alternation of the dipolar energy and, thus, the degree of frustration. However, the nonuniform magnetic frustration is not random. The nonuniform geometrical frustration is the second important ingredient for the definition of the magnetic microstructure in quasicrystals.

The exchange coupling in quasicrystals is also different from that of their periodic counterparts. Atoms on quasiperiodic tilings have not only a varying number of neighbors but also several different nearest neighbor distances (Fig. 1). Accordingly, there are several different values of the exchange force which can even change sign. The existence of several exchange constants J can also exert a significant influence on the microstructure of the quasiperiodic magnets.

In summary, it is obvious that the varying number of nearest neighbors, nonisotropic magnetic frustration, and varying J-constants are important for the micromagnetic ordering in quasiperiodic ultrathin films. From the theoretical point of view no general approach has been made up to now.

The aim of the present study is to achieve a general spatially resolved description of the magnetic ordering on the Penrose two-dimensional tiling. Since the Penrose tiling is aperiodic, an analytical description of the

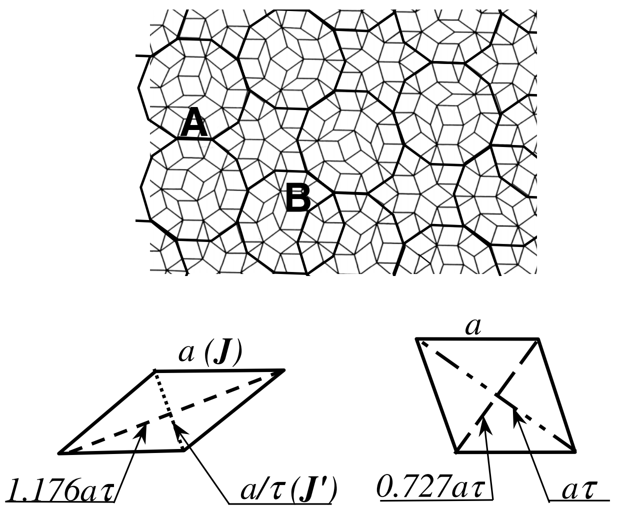

FIG. 1. (a) A section of the Penrose tiling. The original Penrose rhombic tiles and the decagonal tiles are indicated. Two allowed overlapping of decagonal clusters are shown as A and B. (b) The original Penrose rhombic tiles. Five nearest neighbor distances (the sides and the diagonals of the rhombuses) and their lengths are given. τ is the golden mean. The two strongest exchange bonds according to two shortest nearest neighbor distances are denoted as J and J'.

micromagnetic structure is hardly feasible. Therefore Monte Carlo simulations have been performed to find the equilibrium spin configurations at a given temperature. We present as well an original experimental dipolar system made of 309 small magnets on the Penrose tiling. In the Monte Carlo simulations the local ferromagnetic exchange interaction and the long-range dipolar coupling are considered. The experimental system represents a pure dipolar model which corresponds to the numerical simulations for zero exchange interaction. The effects of the indirect exchange coupling are neglected in this study. For a monolayer of three-dimensional vector spins $\mathbf{S}_i$ the Hamiltonian is given by

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_{ij} \left(\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{\mathbf{r}_{ij}^3} - 3 \frac{(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{\mathbf{r}_{ij}^5} \right), \quad (1)$$

where J is the exchange coupling constant and $\langle i, j \rangle$ refers to the nearest neighbors, D the dipolar coupling parameter, and $\mathbf{r}_{ij}$ the vector between sites i and j.

The simulations have been carried out on finite Penrose tilings with free boundary conditions. The samples are squares or rectangles containing 400, 2500, and 10 500 magnetic moments. We have also used circular areas to cross-check our results. We have considered the dipolar interaction of each magnetic moment with all the other moments; i.e., we did not use a cutoff in calculating the dipolar coupling. The Monte Carlo procedure is the same as described in a previous publication [6]. The experimental model concerns a 480 mm $\times$ 480 mm Penrose tiling of magnets of 4 mm length separated by 30 mm. The large distance between the magnets is chosen on purpose to minimize multipolar terms that can trap the system into metastable states [7]. The magnets are put onto nonmagnetic vertical axes and can rotate in the x, y plane.

In order to calculate the exchange energy the set of nearest neighbors that are coupled via the short-range interaction has to be defined. In periodic crystals the exchange coupling between next nearest neighbors is usually enough to ensure the magnetic order. In quasicrystals the situation is different. The pattern consists of two rhombuses with edges of equal length a, one with angles of 36° and 144° and the other with angles 72° and 108° (Fig. 1). The rhombic tiles are arranged without gaps or overlaps according to matching rules [5]. The smallest distance between neighbor sites is the short diagonal of the tight rhombus (Fig. 1). The exchange interaction along this diagonal J' is nonpercolating; i.e., it can connect the spins into only very small clusters of a maximal size equal to three moments (see Fig. 1). Thus, it cannot ensure the magnetic alignment of the whole sample. To get a long-range magnetic order the exchange coupling along the sides of the rhombuses J must be included (Fig. 1). Usually, in theoretical studies of critical behavior of quasiperiodic systems only J or J and J' interactions are considered (longer bonds are neglected). With such a treatment of bonds the lattice deviates from the original Penrose tiling. In our study five different values of the exchange constant, i.e., for the sides and all diagonals of the rhombuses, have been considered. J has been taken to be unity. The exchange interaction decreases exponentially with the distance between magnetic moments. The strength of the exchange interaction is defined as $J_{ij} = J \exp(1 - \rho_{ij})$, where $\rho_{ij} = r_{ij}/a$ is the distance between two neighboring moments normalized to the length of the side of the rhombuses a. ρ_{ij} takes the lengths of the diagonals of the Penrose rhombuses. The shortest diagonal has a length of $\rho_{ij} = \tau^{-1} < 1$ with τ as the golden mean. Therefore $J' = J \exp(1 - \tau^{-1})$; i.e., J' is larger than J. Further interactions become weaker than J with increasing distance as in that case $\rho_{ij} > 1$.

Magnetic ordering depends on the ratio of exchange to dipolar constant $R = J/D$ and on the radius of the cutoff in the exchange coupling (ρ). We have performed calculations for R varying between 0 ($J = 0$, pure dipolar interactions) and 1000. The cutoff radius in the exchange interaction can take one of four values: $\rho = a$, which means that the exchange coupling is considered only along sides and the shortest diagonal of the Penrose rhombuses, $\rho = 0.727a\tau$, $\rho = a\tau$, or $\rho = 1.176a\tau$. The latter distances correspond to the exchange coupling along the longer diagonals (see Fig. 1).

Figure 2 shows examples of relaxed micromagnetic configurations for pure dipolar interactions obtained

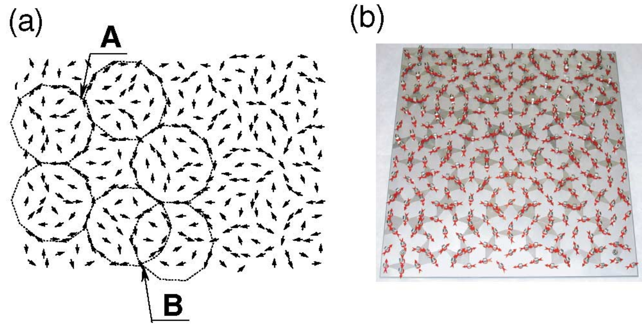

FIG. 2 (color). (a) Monte Carlo simulations. Top view of the portion of the magnetic microstructure in a sample of finite size for pure dipolar interaction, i.e., $R = J/D = 0$. The microstructure has been obtained for a square sample of about 10 500 vector spins on the Penrose tiling for $D/k_BT = 100$. The spins belonging to the perimeter of decagons (marked) form closed chains. The chains overlap according to rules given in Fig. 1. (b) Experimental model. The perspective view of the magnetic microstructure. The red arrows represent the orientation of dipolar moments of magnets fixed onto the nodes of the Penrose tiling (rhombuses). The magnets can rotate in the horizontal plane.

in the numerical [Fig. 2(a)] and in the experimental [Fig. 2(b)] models. Both studies show that after different relaxation procedures a micromagnetic pattern can have a different local arrangement of dipoles. The total energy, however, is always identical. Thus, the ground state in the case of $J = 0$ is highly degenerate. All patterns, theoretical and experimental, have features in common. Magnetic moments are ordered in circular loops. The diameters of the loops are identical all over the sample. The loops overlap. This overlapping is not accidental but follows certain rules. Amazingly, these rules coincide with the recently proposed "decagonal model" of quasicrystals [8–11].

In 1991 it was realized [8] that the planar Penrose tiling can be generated using a single kind of tile, a decagon. Every decagon consists of Penrose rhombuses. In contrast to the conventional tiling description the decagonal atomic clusters overlap, which means that they share atoms with their neighbors. The overlapping rules have been mathematically proven [9]. Only two types of the overlap (A and B) are allowed [8]. Location of "A" and "B" in a Penrose tiling are marked in Figs. 1 and 2(a).

The decagons can be easily recognized in the magnetic microstructure [Figs. 2(a) and 2(b)]. In order to minimize the dipolar energy the magnetic moments located on the perimeter of a decagon form closed chains. The moments are coplanar to the sides of the decagons. The overlapping rings of magnetic moments can have the same or opposite sense of rotation. The orientation of the moments that do not belong to the perimeter of decagons is highly frustrated and varies from cluster to cluster. The overlapping magnetic decagon chains form a quasiperiodic pattern. In case of pure dipolar interaction the magnetic pattern is formed on the scale of the tiling constant; i.e., a microscopic pattern is formed. In zero magnetic field this state is degenerate and represents a manifold of quasiperiodic spin configurations. All frustrated systems that have been investigated have either a continuously degenerated, periodic ground state (spins on a honeycomb, a kagome, a triangular, a pyrochlore lattice [12]) or a completely disordered one (spin glasses). The superposition of both types of frustration has not been reported yet. Thus, a magnetic system on a Penrose tiling belongs to a new class of frustrated systems where the degenerated ground state is aperiodic and consists of two parts: ordered decagon rings and disordered spin-glass-like phase inside the decagons.

In the following we will discuss the situation where the exchange coupling is switched on. In the quasiperiodic Penrose tiling with high R, i.e., with the strong exchange interaction, we find a single domain for all cutoff radii $\rho \geq a$. It means that the exchange coupling acting along the two shortest bonds (J and J') is enough to ensure the ferromagnetic order. However, the degree of magnetic order increases with increasing ρ. While the low temperature magnetization is unity for the large exchange cutoff radius $\rho = 1.176a\tau$, it is $\bar{M} = 0.975$ for $\rho = a$ ($R = 10^3$). Hence, the ferromagnetic order in quasicrystals depends on the cutoff radius taken for the exchange interaction. This can cause strong inhomogeneities of the magnetization at the boundaries of laterally confined magnet with quasiperiodic structure.

In finite samples on square and triangular lattices single domain configurations have been found for high R values while in-plane vortex structures dominate for $R \approx 1$ [13]. The vortex phase arises as a result of the influence of the sample boundaries. The dipolar interaction prefers to keep the magnetic moments in the film plane and parallel to the sample edges to avoid formation of magnetic poles. The exchange energy cares for the parallel orientation of the neighboring moments. The interplay of the different contributions leads to formation of the vortex structure with dimensions of the sample size. For the Penrose tiling the situation is completely different. For all R-ratio and cutoff radii the macroscopic vortex configuration is energetically unfavorable with regard to the exchange interaction. When the dipolar energy becomes strong enough to compete with the exchange energy ($R < 0.5$) the microscopic decagonal pattern starts to form (Fig. 3). The decagonal pattern differs from that of the pure dipolar case when exchange interaction is effective. The strong exchange coupling lifts the degeneracy of the decagonal magnetization configuration found for $J = 0$. Magnetic moments are nearly coplanar with the sides of the decagons as in the pure dipolar case. The average magnetization, however, is not zero; i.e., the magnetic moments have some preferential direction [Fig. 3(a)]. We call such magnetization configuration quasiferromagnetic decagonal structure. A

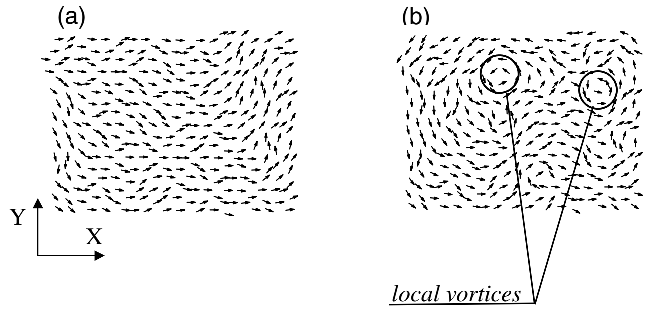

FIG. 3. (a) Top view of the portion of the quasiferromagnetic spin configuration in a sample of finite size for $\rho = 1.176a\tau$ and $R = J/D = 5$. The magnetic moments are nearly coplanar to the sides of the decagons. The X component of the average magnetization is $M_X = 0.85$. (b) An example of a planar spin configuration in the region of transition from the single domain to the decagonal structure for $\rho = 1.176a\tau$ and $R = J/D = 0.4$. The microstructures have been obtained for square and disk-shaped samples of 400 and 10 500 magnetic moments at $J/(k_B T) = 100$. The magnetic moments at the edges are oriented mainly parallel to the boundary as in a conventional vortex structure. However, only local vortices inside the decagons exist.

further decrease of the ratio R leads to an increasing influence of the dipolar interaction on the magnetic microstructure. To minimize the magnetostatic energy the dipoles form lines at the edges of the sample as in conventional vortex structure [Fig. 3(b)]. However, a macroscopic vortex does not form for any shape of the sample. Small local vortices can appear only inside some decagon rings [Fig. 3(b)].

Thus, the influence of the boundaries does not lead to the formation of a macroscopic vortex in a Penrose tiling. The reason for this phenomenon is the spatial variation of the number of nearest neighbors and the exchange interaction strength. As the strength of the exchange interaction decreases exponentially with the distance, J is much stronger for neighbors with $\rho_{ij} \leq a$, i.e., with $J \geq 1$, than for neighbors with $\rho_{ij} > a$. The magnetic moments with $\rho_{ij} \leq a$ are situated mainly on the perimeter of the decagons. It is energetically more preferable to keep these moments parallel than the other ones which causes the appearance of decagonal chains and the local vortices. The formation of macroscopic configurations is suppressed in favor of the microscopic quasiferromagnetic pattern.

In conclusion, the stable magnetization configurations of magnets on a quasiperiodic tiling have been derived theoretically. In contrast to periodic lattices, the formation of macroscopic vortex configuration is suppressed in favor of the microscopic quasiferromagnetic pattern. For low R ratios a new microscopic structure, the quasiferromagnetic decagonal pattern, represents the minimum of the free energy. For pure dipolar interaction the decagonal pattern represents a new class of frustrated systems where the degenerated ground state is aperiodic and consists of two parts: ordered decagon chains and disordered spin-glass-like phase inside the decagons.

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[1] See for a review H. P. Oepen and J. Kirschner, Curr. Opin. Solid State Mater. Sci. **4**, 217 (1999).
[2] Z. Luo, S. Zhang, Y. Tang, and D. Zhao, Scr. Metall. Mater. **28**, 1513 (1993); A. Niikura, A. P. Tsai, A. Inoue, and T. Masumoto, Philos. Mag. Lett. **69**, 351 (1994); A. P. Tsai, A. Niikura, A. Inoue, T. Masumoto, Y. Nishida, K. Tsuda, and M. Tanaka, Philos. Mag. Lett. **70**, 169 (1994).
[3] See for a review U. Grimm and M. Baake, in *The Mathematics of Long-Range Aperiodic Order*, edited by R.V. Moody (Kluwer, Dordrecht, 1997), p. 199.
[4] See for a review K. De'Bell, A. B. MacIsaac, and J. P. Whitehead, Rev. Mod. Phys. **72**, 225 (2000).
[5] R. Penrose, Bull. Inst. Math. Appl. **10**, 266 (1974).
[6] E. Y. Vedmedenko, H. P. Oepen, A. Ghazali, J.-C. S. Lévy, and J. Kirschner, Phys. Rev. Lett. **84**, 5884 (2000).
[7] E. Olive and P. Molho, Phys. Rev. B **58**, 9238 (1998).
[8] S. E. Burkov, Phys. Rev. Lett. **67**, 614 (1991).
[9] P. Gummelt, in *Proceedings of the 5th International Conference on Quasicrystals*, edited by C. Janot and R. Mosseri (World Scientific, Singapore, 1995), p. 84.
[10] P. J. Steinhardt and H. C. Jeong, Nature (London) **382**, 433 (1996); H. C. Jeong and P. J. Steinhardt, Phys. Rev. B **55**, 3520 (1997).
[11] K.W. Urban, Nature (London) **396**, 14 (1998).
[12] P. Schiffer, Nature (London) **420**, 35 (2002).
[13] E. Y. Vedmedenko, A. Ghazali, and J.-C. S. Lévy, Phys. Rev. B **59**, 3329 (1999); J. Sasaki and F. Matsubara, J. Phys. Soc. Jpn. **66**, 2138 (1996).