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Periodic average structures of colloidal quasicrystals

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We experimentally study the phase behaviour of a charge-stabilized two-dimensional colloidal monolayer which is subjected to a one-dimensional quasiperiodic substrate potential. Upon increasing the laser intensity, we observe a transition from a periodic to a quasiperiodic state. It proceeds via the formation of an intermediate periodic average structure (PAS) which is related to the quasiperiodic lattice by a bounded 1-1 mapping. Because PAS can transform to crystals and quasicrystals by minute particle displacements, they provide a mechanism to allow for interesting insights into the relationship between periodic and quasiperiodic order.

1 Introduction

Although *periodic* crystals and quasicrystals, both exhibit long-range positional order, their atomic configuration is markedly different[1]: In contrast to a periodic arrangement, quasicrystals are created by the aperiodic distribution of atoms[2–4]. In view of these striking differences regarding their intrinsic order, the experimental observation of phase transitions between quasicrystals and crystals (or their corresponding approximants) upon variations in temperature[5–7] or pressure[8], exposure to electron beams[9,10] or small changes in the sample conditions[11], is surprising. In particular, it is not obvious what atomic movements are required to allow for such phase transitions. Periodic average structures (PAS) have been suggested as a possible solution to this problem because they are related to quasicrystals by a 1-1 mapping[12–14] and can provide an explanation how atoms can change between periodic and quasiperiodic positions by minute (smaller than the smallest atomic bond length) particle displacements[15]. Although numerical comparison between measured and calculated diffraction patterns provides some evidence for the occurence of PAS[5,15], periodic average structures have not yet been observed in real space experiments.

Here, we experimentally demonstrate the transition between a PAS and a quasicrystal in a colloidal system under real space conditions. Such soft quasicrystals have recently received considerable attention as model systems to understand the stability and structure of quasicrystals[16–20]. In contrast to atomic systems, where quasiperiodic order is the sole result of the interatomic forces, here quasiperiodic order is imposed by an external potential acting on the colloidal particles. This allows the use of systems interacting with simple and isotropic pair potentials and thus to study the generic conditions under which such phase transitions can occur. Since most theoretical studies on PAS have been performed in one-dimensional systems[13,14], in our experiments we consider a colloidal monolayer which is subjected to a one-dimensional quasiperiodic substrate potential.

2 Experimental materials and methods

Our experiments were performed with an aqueous suspension of highly charged polystyrene spheres with diameter $\sigma = 3.9\,\mu$m which were functionalized with -COOH groups and suspended in water (Microparticles GmbH, Berlin). Gravity localizes the particles about 100 nm above the negatively charged bottom plate of the sample cell, where they form a two-dimensional monolayer with out-of-plane fluctuations less than 5% of the particle diameter. The particles interact via a screened Coulomb potential $u(r) = A \cdot \exp(-\kappa r)/r$. This type of pair potential[21] has been demonstrated to be also applicable when the particles are confined close to a wall[22,23]. The prefactor A and the Debye screening length κ^{-1} have been determined by comparison of pair correlation functions which have been obtained experimentally and by two-dimensional numerical simulations[24]. From this we obtain $A \approx 10^{10}\ k_B T \cdot \mu$m and $\kappa^{-1} = 360 \pm 40\,$nm, respectively. To achieve such large screening lengths ions were removed from the sample by pumping the suspension through a circuit containing an ion exchange resin[25].

Substrate potentials with one-dimensional quasiperiodic order were created by a scanned optical line tweezer[26] which interacts with the particles due to optical gradient forces[27] and thus serves as a substrate potential[28] whose strength V_0 is set by the laser intensity I. The patterns were formed by reflec-

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tion of a laser beam of a frequency doubled Nd:YAG laser ($\lambda = 532\,\mathrm{nm}$) on a single-axis galvanostatically driven mirror (Scanlab) whose position was controlled by the voltage of a programmable frequency generator. Afterwards the circular cross section of the beam is expanded to a line of 3 mm length and half width $w = 2.5\,\mu\mathrm{m}$.

Line patterns comprised of 40 parallel lines were achieved by application of appropriate signals to the mirror which result in a quasiperiodic step-wise motion of the laser line. The time duration for an entire mirror cycle was set to $T = 0.025$ s, which ensures, that the light pattern can be considered as a quasi-static potential for the colloidal particles[26]. In our experiments, we have chosen patterns corresponding to a Dodecanacci series SLLLSLLSLL... which is formed by the quasiperiodic sequence of two length scales L and S with $\mathrm{L/S} = \tau_{\mathrm{Dod}} = (1+\sqrt{3})/2$. For further information about the Dodecanacci model and other 1D quasiperiodic sequences we refer to the literature[29,30]. Particle positions were recorded via digital video microscopy at frame rates of 1 Hz and determined with an accuracy of about ± 50 nm by digital video microscopy. Two-dimensional particle density distributions and pair correlation functions were obtained by averaging over 1000 frames. The substrate potential is obtained from the probability distribution of a highly diluted colloidal suspension. Under equilibrium conditions, this distribution is given by the Boltzmann distribution from which the shape and the depth of the quasiperiodic substrate potential are obtained.

3 Results and discussions

3.1 Crystal to quasicrystal transition

Fig. 1 shows the particle density distribution of a colloidal monolayer in the presence of a dodecagonal line potential with $\mathrm{L} = 8.0\,\mu\mathrm{m}$ and $\mathrm{S} = 5.9\,\mu\mathrm{m}$ for increasing substrate potential strength. Particle positions have been determined by digital video microscopy (Methods). The experiment has been performed at a mean particle distance of $a = 7.2\,\mu\mathrm{m}$ ($\rho = 0.018\,\mu\mathrm{m}^{-2}$). At such particle distances, a liquid state is observed for $V_0 = 0\ k_{\mathrm{B}}T$ (Fig. 1(a)).

For $V_0 = 1.2\ k_{\mathrm{B}}T$, the particles arrange in strings along the y-direction, i.e. parallel to the imposed quasiperiodic light potential (Fig. 1(b)). It should be realized, however, that the colloidal strings do not exactly follow the quasiperiodic substrate potential but are laterally shifted by $0.5-1.5\,\mu\mathrm{m}$ (corresponding to about 20% of the mean particle distance) relative to the substrate potential. These shifts are visualized by imposing the potential minima as red lines on the figure. Clearly, the colloids form a string pattern with *periodic* spacing with lattice constant $d \approx 7.5\,\mu\mathrm{m}$. Only when V_0 is increased to a much higher value, i.e. $16\ k_{\mathrm{B}}T$, the colloids resemble the quasiperiodic substrate pattern and form a one-dimensional quasicrystal (Fig. 1(c)).

3.2 characterization of 1D periodic structures

To understand the formation of a periodic colloidal structure in the presence of a *quasiperiodic* substrate potential, we first investigated how the spacing between colloidal strings depends on the particle number density ρ. During these experiments, the potential strength was kept at $V_0 = 1.5\ k_{\mathrm{B}}T$. Fig. 2 shows probability distributions $p(d)$ of the particle's lateral next neighbour distance d for mean particle distances $6.5\,\mu\mathrm{m} < a < 8.2\,\mu\mathrm{m}$ or densities $0.011\,\mu\mathrm{m}^{-2} < \rho < 0.022\,\mu\mathrm{m}^{-2}$.

Despite some broadening of the distributions, the maxima remain at $\bar{d} = 7.5 \pm 0.1\,\mu\mathrm{m}$, independent of the particle density. This suggests, that the emerging periodic structure is not the result of particle interactions only. As will be shown in the following, the lattice constant of the observed structures is in quantiative agreement with the PAS corresponding to the quasiperiodic substrate potential.

3.3 Theoretical approach for constructing Periodic Average Structures (PAS) of quasicrystals

Generally, quasicrystalline structures can be obtained by a lower-dimensional section of a higher-dimensional periodic crystal[31–33]. However, when such hypercrystals are projected under an oblique angle onto the physical space, this yields a discrete *periodic* structure, i.e. the periodic average structure, with lattice constant P_{av} [14] (Fig. 3).

3.3.1 Periodic average structures (PAS) of a Dodecanacci sequence

The PAS is a modification of the higher-dimensional description of incommensurately modulated structures (IMS) introduced by Janner, Janssen, and de Wolff in the 1970s[31,32]. IMS have an aperiodic diffraction diagram with satellite peaks which can be embedded in a periodic higher-dimensional space. On transformation to physical space this leads to a periodic average structure, produced by vertical projection of oblique acceptance domains which determine the position of the atoms[34,35]. On the other hand, the acceptance domains lead to an aperiodic sequence when cut with physical space. A shearing of the embedding space parallel to the physical space such that the acceptance domains become vertical (red bars in Fig. 3) leads to the standard PAS picture for quasicrystals. Now the higher-dimensional bands of the PAS are oblique (green bars). In the simple case of a one-dimensional sequence embedded periodically in two dimensional space, a square lattice is used as the support of the acceptance domains. In that case, the frequency of the tiles and their length fraction has to be equal. For the one-dimensional quasiperiodic structure discussed here and described in Fig. 3, this requires a rectangular

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lattice (See also Socolar *et. al.*[33] for the general case). The basis vector $\mathbf{d}_1$ has to be $\sqrt{2}$ longer than the basis vector $\mathbf{d}_2$, and the fraction of $N_L/N_S : L/S$ is 2. The length of the acceptance domains can be chosen as $N_L + N_S$, and the length of the tiles as $L\sqrt{2}$ and $S\sqrt{2}$. The inclination angle of the supporting rectangular lattice with respect to the physical space must be then $\phi = 27.368°$ or $arctan(\sqrt{2}/(\sqrt{3}+1))$.

Because the resulting PAS corresponds to a bounded 1-1 mapping onto the quasicrystalline structure[12], it provides a natural link between structures having periodic and quasiperiodic order. The construction of a PAS for a Dodecanacci sequence is schematically illustrated in Fig. 3 and demonstrates how the transition between both structures is achieved by local and minute particle displacements[10,14,15,36]. The unique mapping between the quasiperiodic sequence and the periodic average structure requires, that the number of vertices remain identical. Therefore, $(N_L + N_S)P_{av} = N_L L + N_S S$ with N_L and N_S the frequency of L and S segments, respectively[14]. Because for a Dodecanacci sequence $N_L : N_S = 2\tau_{Dod}$[36], one obtains

$$P_{av} = 2S(2 - \tau_{Dod}). \quad (1)$$

3.4 Experimental verification of Periodic Average Structures of quasicrystals

For $S = 5.9 \pm 0.1\,\mu m$ this leads to $P_{av} = 7.48 \pm 0.13\,\mu m$ which is in excellent agreement with the lattice constant $\bar{d}$ observed in our experiments (Fig. 1(b), Fig. 2). Qualitatively, PAS formation in our experiments is the result of the combination of particle-particle and particle substrate interactions. While the pair interaction favors a uniform spacing, the light field aims to impose quasiperiodic order on the colloidal strings. Accordingly, the PAS can be considered as an intermediate phase since it requires only minimal particle displacements relative to a perfect periodic or quasiperiodic lattice. As a result, the width of the particle distance distributions $p(d)$ in a PAS should be somewhat larger than that in a conventional crystal. In addition, the symmetry of $p(d)$ will be different as shown in Fig. 4. While distance histograms of entirely electrostatically-stabilized colloidal crystals are always strongly asymmetric and fall off more rapidly towards smaller distances due to the exponential distance dependence of $u(r)$ (Fig. 4b), this is not the case in the histograms of the PAS (Fig. 4a and Fig. 2). This is because particle distances larger than L do not occur in PAS leading to a sharp cutoff in $p(d)$ towards larger distances. As a result, the skewness of the distance histograms of crystals and PAS are opposite.

The complete phase behavior of a colloidal monolayer on a one-dimensional quasiperiodic substrate as a function density ρ and the mean particle distance a is shown in Fig. 5(a). To distinguish different phases, we have used $\bar{d}$ as the order parameter. For $a \leq 6.0\,\mu m$ ($\rho \geq 0.027\,\mu m^{-2}$), the particle pair interaction is more than 50 times stronger than the substrate potential. Under such conditions, the quasiperiodic light field provides only a small perturbation and we observe an undistorted two-dimensional triangular crystal with lattice constant a which is aligned along the y-direction. Accordingly, the spacing between colloidal strings is $\bar{d} = \frac{\sqrt{3}}{2}a$ in agreement with our data (green symbols, dashed line). In the range $6.5\,\mu m < a < 8.2\,\mu m$, i.e $0.011\,\mu m^{-2} < \rho < 0.022\,\mu m^{-2}$, the line spacing remains rather constant around $\bar{d} = 7.47\,\mu m$ (red symbols). As already mentioned, this value agrees with the periodicity of the PAS obtained from Eq. (1) (horizontal line). Further increase of the particle distance to $a > 8.2\,\mu m$ finally leads to the dominance of the substrate potential and the colloids follow the quasiperiodic light potential pattern (blue symbols). Then, the histogram $p(d)$ exhibits a double-peaked structure with the peak positions approaching L and S for increasing mean particle distance. The transition between the crystal and the quasicrystal appears to be rather smooth which suggests a continuous phase transition which is consistent with the absence of hysteresis effects in our experiments. To demonstrate the robustness of PAS against variations in the length scale of the Dodecanacci sequence, we also varied L and S with $L/S = (1+\sqrt{3})/2$ as before. This is shown in Fig. 5b where we plotted $\bar{d}$ vs. S for $a = 7.4\,\mu m$ ($\rho = 0.017\,\mu m^{-2}$) and $V_0 = 1.5\ k_B T$. The measured data (symbols) are in excellent agreement with Eq.(1) (solid line).

Periodic average structures are also predicted for other quasiperiodic symmetries[14,36,37]. Therefore, we performed experiments where the laser pattern was modulated according to a Fibonacci sequence (LSLLSL,...). Here, the ratio of the length scales L/S and that of the corresponding frequencies $N_L : N_S$ is given by the golden mean $\tau_{Fib} = (1+\sqrt{5})/2$. Accordingly, the periodicity of the associated PAS is given by $P_{av} = (3 - \tau_{Fib})S$[14]. Fig. 6 shows the particle density distribution of a colloidal monolayer with $a = 7.0\,\mu m$ ($\rho = 0.02\,\mu m^{-2}$) when exposed to a Fibonacci light pattern with $V_0 = 1.5\ k_B T$ and $L = 8.7\,\mu m$. Again, the colloids arrange in a periodic one-dimensional pattern with spacing $\bar{d} = 7.4\,\mu m$ which is in good agreement with the periodicity of the corresponding periodic average structure $P_{av} = 7.46 \pm 0.14\,\mu m$. Due to the larger L/S ratio, the formation of PAS requires larger relative particle displacements than in the case of a Dodecanacci sequence[38]. Accordingly, the range of particle densities, where the PAS is observed is much smaller under such conditions.

Although, PAS can arise for different types of line sequences, it should be realized, that it is not possible to find a PAS for any arbitrary lattice. This is easily seen by considering the example of a line distribution of two length scales L and S where $L = 3S$ and the corresponding frequencies N_L and N_S are equal. Since the number of vertices must be identical (1-1 mapping), in case there exists a PAS for this sequence,

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its periodicity must be $P_{av} = 2S$. This, however, would require particle shifts of $P_{av} - S = S$ which is impossible since the quasiperiodic ordering requires a bounded fluctuation of the particles about their positions. For more details see the discussions made by Aubry *et al.* [39].

The concept of PAS finds also use in higher-dimensional quasicrystalline structures [12,14] which can be experimentally achieved e.g. by interference of five laser beams [18]. Because in two-dimensional systems there is no simple 1-1 mapping between periodic and quasiperiodic structures [36], colloidal experimental studies will provide direct information how crystal to quasicrystal transitions are achieved under such conditions. Since the formation of PAS only requires small particle displacements, they may also form during atomic hetero-epitactic monolayers on quasicrystalline surfaces to minimize elastic strain with the substrate. Such monolayers are currently considered for the development of new materials and devices [38,40].

Finally, it should be mentioned, that in addition to the phases observed here, other structures can form. For example, at higher particle concentrations and smaller Debye screening lengths, phases with interstitial particle strings between the quasiperiodic lines should be observed.

4 Summary

In summary, our experiments demonstrate that a smooth transition between one-dimensional crystals to quasicrystals is possible by the formation of a periodic average structure which is remarkably stable with respect to parameter variations. Obviously, the formation of PAS does not require complex or even anisotropic pair potentials as this is typically encountered in atomic quasicrystalline systems. Rather PAS can be generally expected in situations where periodic and quasiperiodic ordering principles compete against each other.

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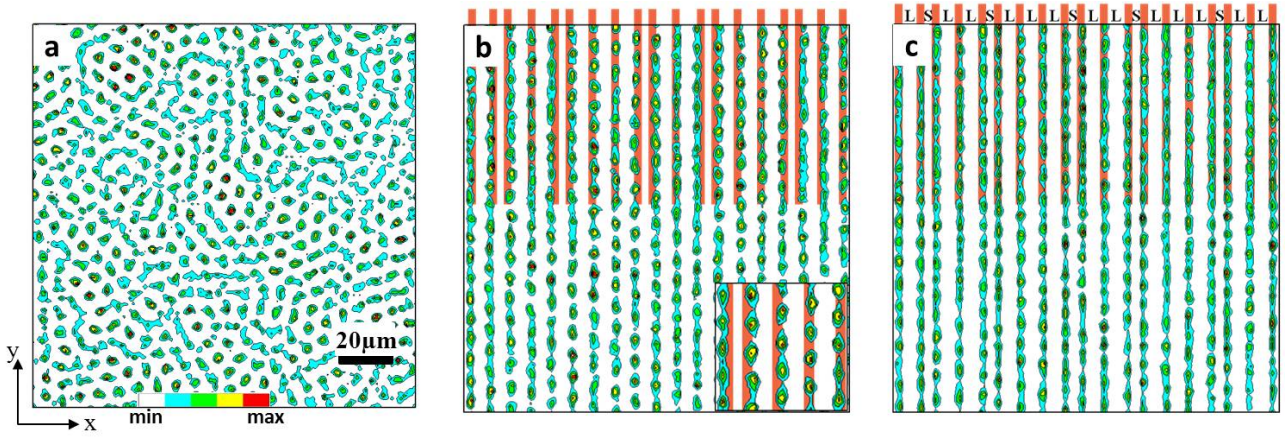

Fig. 1 Contour plots of the two-dimensional average colloidal density distribution in presence of a one-dimensional quasiperiodic substrate potential (L = 8.0 µm, S = 5.9 µm) and for increasing strength (a) $V_0 = 0\ k_BT$: isotropic liquid, (b) $V_0 = 1.2\ k_BT$: one-dimensional periodic crystal with lattice constant $d \approx 7.5$ µm. The inset corresponds to a magnified area and shows how the colloidal strings are shifted in x-direction relative to the quasiperiodic substrate potential (c) $V_0 = 16\ k_BT$: one-dimensional quasicrystal. The vertical red lines correspond to the potential minima of the substrate potential (for clarity, those lines are only shown in the upper part of the figures).

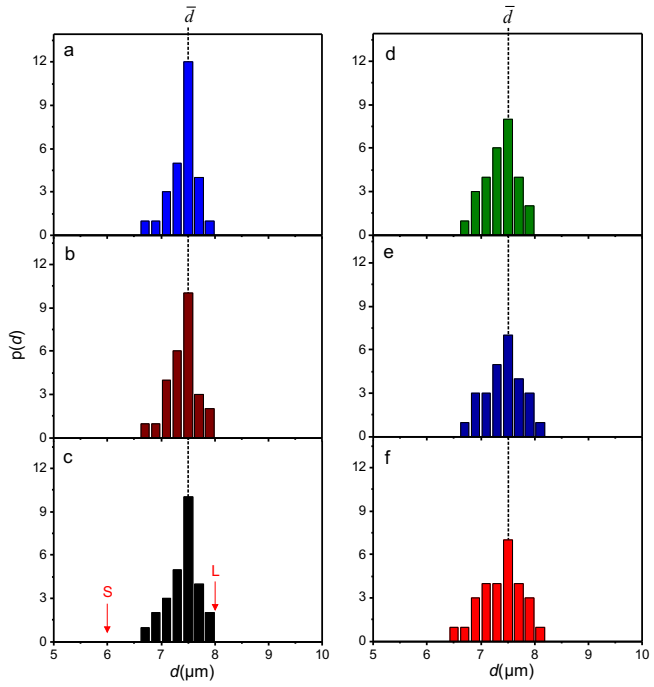

Fig. 2 Measured lateral distance distributions between particles in neighbouring colloidal strings for increasing mean particle distance (a) $a = 6.5$ µm, (b) 6.8 µm, (c) 7.2 µm , (d) 7.6 µm, (e) 8.0 µm to (f) 8.2 µm. The quasiperiodic light potential is set to $V_0 = 1.5\ k_BT$ with L = 8.0 µm and S = 5.9 µm. Despite broadening of the distributions, the maxima, i.e. the lattice constant of the one-dimensional colloidal crystals, remains identical at $\bar{d} \approx 7.5$ µm.

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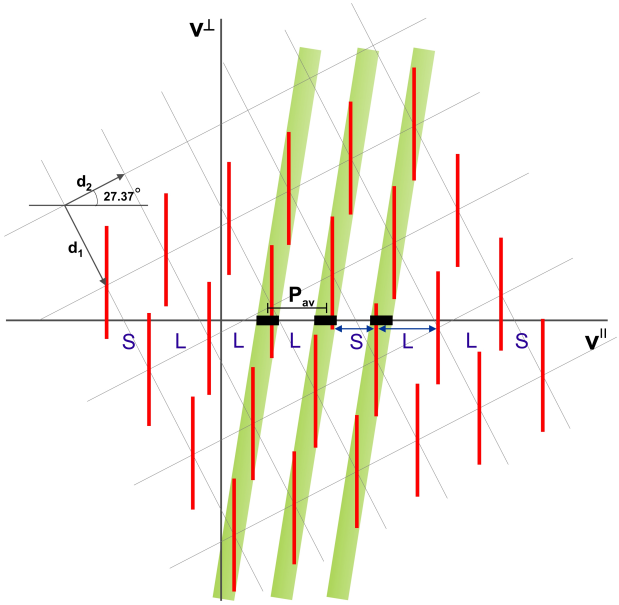

Fig. 3 The 2D hyperspace $(V^{\parallel},V^{\perp})$ is spanned by the unit vectors $\mathbf{d}_1$ and $\mathbf{d}_2$ that define a rectangular unit cell which is rotated by $\phi = \sqrt{2}/(\sqrt{3}+1) \approx 27.37°$. The vector $\mathbf{d}_1$ is a factor $\sqrt{2}$ longer than $\mathbf{d}_2$. The Dodecanacci sequence is obtained by the intersection points of the red vertical bars (hyper-atoms) and the $V^{\parallel}$-axis. When the hyperatoms are projected in oblique direction on the $V^{\parallel}$-axis, as shown by green bars, this leads to periodically spaced regions (black bars) which constitute the corresponding one-dimensional periodic average structures with periodicity P_{av}.

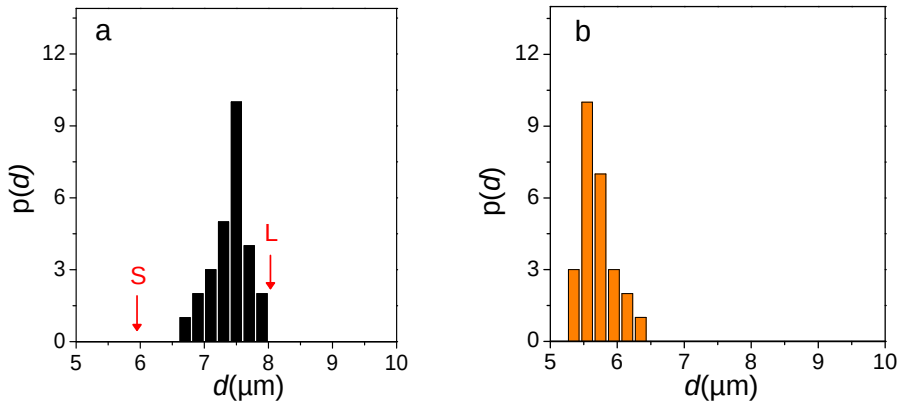

Fig. 4 Comparison of $p(d)$ of a PAS for L = 8.0 μm, S = 5.9 μm, a = 7.2 μm shown in a) (see also Fig. 2(c)), and a conventional two-dimensional colloidal crystal b), the later formed at smaller mean particle distance a = 6.0 μm in the absence of a quasiperiodic substrate potential. Both structures are formed at $V_0 = 1.5\ k_B T$.

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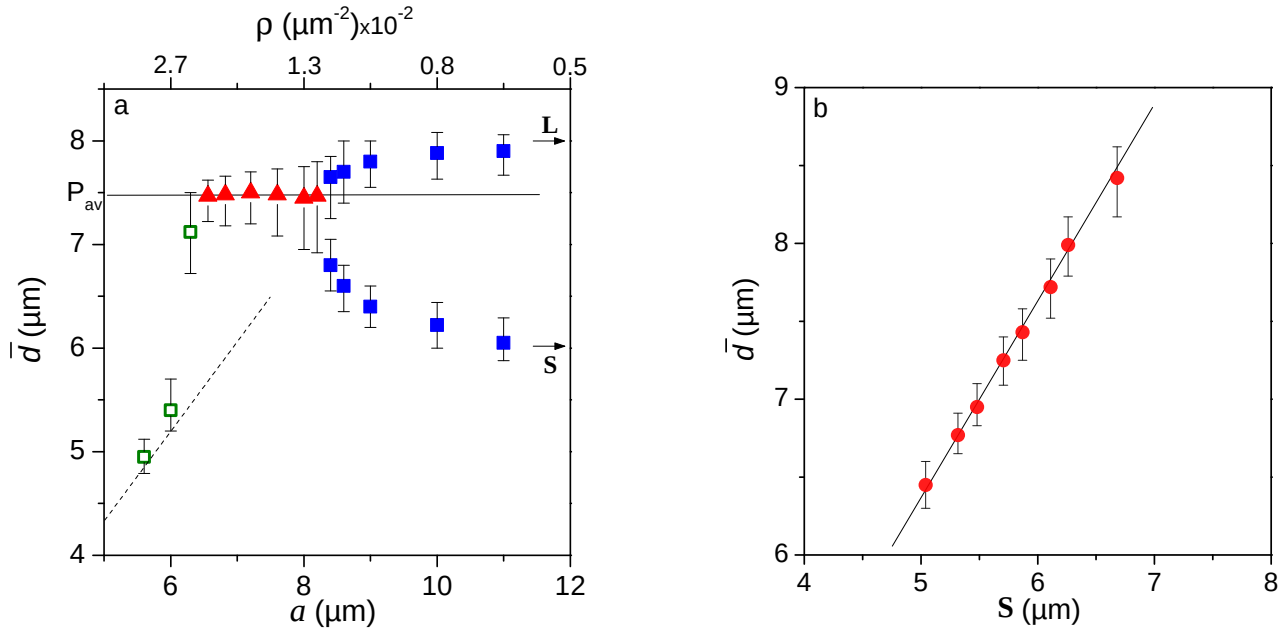

Fig. 5 a) Line distance $\bar{d}$ as a function of density ρ and the mean particle distance a. The parameters of the quasiperiodic substrate are $V_0 = 1.5 k_B T$, L = 8.0 μm and S = 5.9 μm, respectively. With increasing particle distance (or decreasing the density) we find a triangular crystal (green squares, dashed line), a one-dimensional PAS (red triangles) and a one-dimensional quasicrystal (blue squares). The horizontal line corresponds to the predicted lattice constant of the PAS. b) Experimentally determined periodicity $\bar{d}$ of the PAS with $a = 7.4\,\mu m$ ($\rho = 0.017\,\mu m^{-2}$) as a function of the absolute length scale of the quasiperiodic sequence (red circles). The solid line is the prediction according to Eq.(1). The error bars correspond to the halfwidth of the line distance histograms.

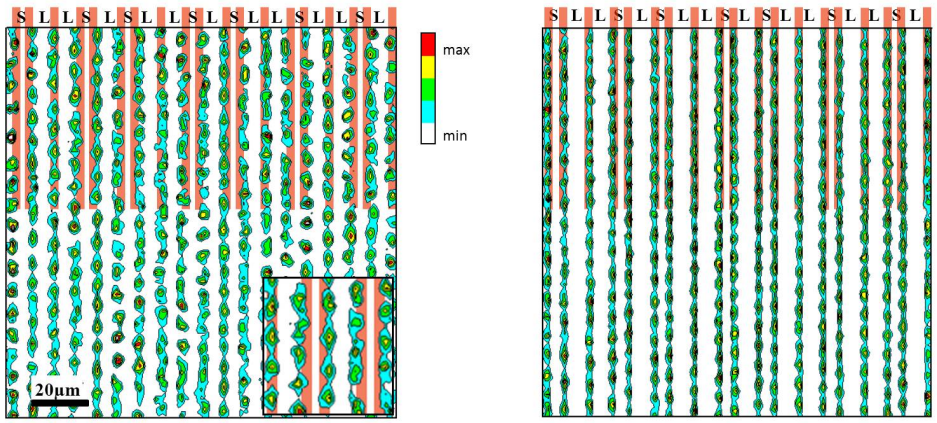

Fig. 6 Contour plot of the colloidal particle density in the presence of a Fibonacci sequence (vertical red lines) with L = 8.7 μm and potential depth a) $V_0 = 1.5\ k_B T$ where the particles rather form colloidal lines with periodic spacing 7.4 μm which is in quantitative agreement with the periodicity of the PAS related to the Fibonacci sequence. b) V_0=16 $k_B T$, the particles follow the potential wells and form a quasiperiodic sequence. The inset in a) shows a magnified area.

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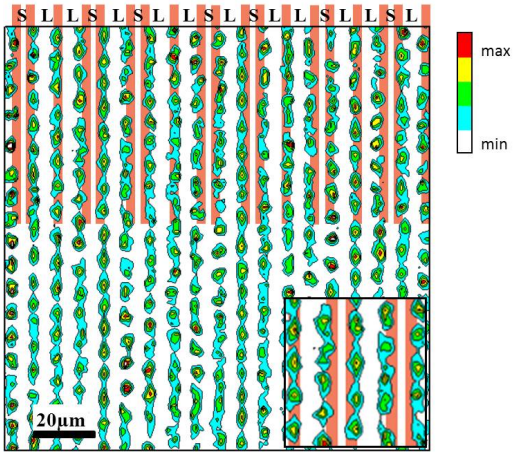
Colour graphic



Observation of periodic average structure of a colloidal monolayer subjected to a one-dimensional quasiperiodic laser potential.

Soft Matter Accepted Manuscript

Dear Editors:

A table of contents entry includes:

1- Revised manuscript entiltled: “Periodic average structures of colloidal quasicrystals” in tex.file with 6 figures embedded at the end.

2- PDF of the revised manuscript with figures in the same file.

3- Colour graphics – One Figure and a statement.

5- Reply letter to the editor.

6- 7 figures in separate files.

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