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Archimedean lattices in the bound states of wave interacting particles

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Abstract – The possible periodic arrangements of droplets bouncing on the surface of a vibrated liquid are investigated. Because of the nature of the interaction through waves, the possible distance of binding of nearest neighbors is multi-valued. For large amplitude of the forcing, the bouncing becomes sub-harmonic and the droplets can have two different phases. This effect increases the possible distances of binding and the formation of various polygonal clusters is observed. From these elements it is possible to assemble crystalline structures related to the Archimedean tilings of the plane, the periodic tesselations which tile uniformly the 2D plane with convex polygons. Eight of the eleven possible configurations are observed. They are stabilized by the coupling of two sub-lattices of droplets of different phase, both contributing to sustain a common wave field.

Introduction. – Periodic structures can result from different ordering principles. During crystallization, the local condensation of atoms having a single distance of binding is responsible for the build-up of macroscopic crystals. In contrast the periodic patterns formed by standing waves result from global resonances of waves having a large coherence length. For this reason they depend on the large-scale shape of the domain in which they grow. These two types of organizations can be observed on a fluid interface, in the rafts formed by floating bubbles [1], and in the wave patterns due to the Faraday instability [2], respectively. Here, as will be shown, we investigate an intermediate case between these two extremes.

It was shown recently [3–5] that droplets bouncing on a vertically vibrated fluid interface could interact and form aggregates. The non-local interaction between drops is due to the damped surface wave they emit. In the self-organized stable arrangement, each drop is at such a position that its successive collisions with the interface leave it motionless. For weak forcing, each drop bounces in the trough formed by the first antinode of the wave generated by its nearest neighbours. This is a Lennard-Jones type of interaction with short-range repulsion and long-range attraction, with a single distance of binding. This interaction leads to the spontaneous formation of crystalline aggregates with a triangular lattice (fig. 1).

However when the forcing is increased, because of the spatial periodicity of the waves, the interacting drops can be stable at several possible distances corresponding to successive antinodes. The experiment reveals the formation of a large variety of patterns involving different polygons. This led us to wonder if this system lent itself to the formation of crystalline arrangements related to the Archimedean tilings. The Archimedean tilings (shown in fig. 2) are the possible uniform tessellations of the two-dimensional Euclidian space with regular polygons. In these structures, first described systematically by Kepler [6], the polygons can be either identical or

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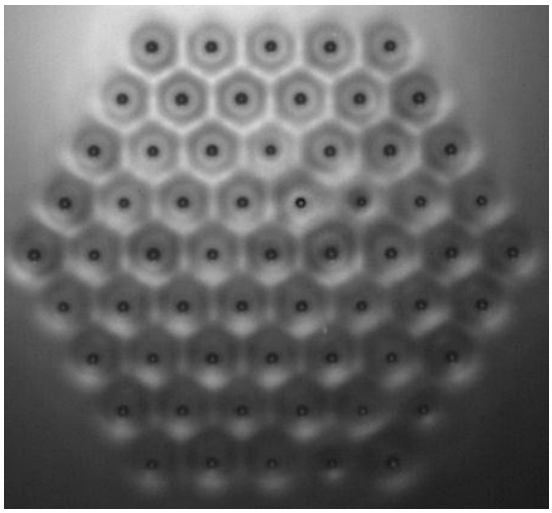

Fig. 1: The crystalline organization of a large number of droplets ($N = 61$) with a triangular lattice.

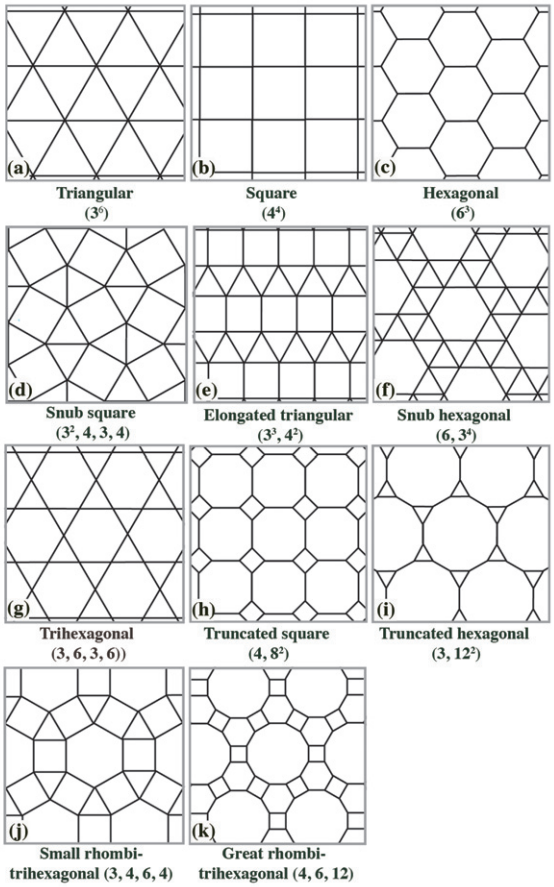

Fig. 2: The 11 Archimedean lattices, ordered by increasing complexity. Three are formed of one single type of polygons: (a) triangles, (b) squares and (c) hexagons. Six are formed of two types of polygons: (d) and (e) triangles and squares, (f) and (g) triangles and hexagons, (h) squares and octagons, (i) triangles and dodecagons. Two are formed of three types of polygons (j) triangles, squares and hexagons, (k) squares, hexagons and dodecagons.

different. The tiling is said to be uniform because all the vertices are identical up to rotation. In the 2D plane, only eleven such tilings exist. Each of them is characterized by the structure of its vertex. The conventional notation in the Grünbaum-Shepard [7] classification gives the relative position of the regular n-sided polygons meeting at the vertex. For instance, in the structure noted $(3^2, 4, 3, 4)$, a vertex is surrounded by two neighbouring triangles, a square, a triangle and a square. The identity of all the links and all the vertices suggests that these tilings could form the structure of crystalline lattices. Such crystals would be of interest since it was theoretically shown that they can have complete photonic band gaps [8]. However, in reality only a few of them were observed [8–13]. Presumably this is due to the fact they do not correspond to dense pilings. Some of the polygons being large (*e.g.* octagons or dodecagons) they form voids in the structure which tend to collapse in order to become more tightly packed.

Experimental set-up. – We study the aggregation of identical droplets bouncing on a vertically vibrated fluid interface. The experiments are performed on a liquid bath of thickness $h_0 = 4$ mm subjected to a vertical oscillating acceleration $\gamma = \gamma_v \cos(2\pi f_0 t)$. The liquid is silicon oil with viscosity $\mu_1 = 20 \cdot 10^{-3}$ Pa · s, surface tension $\sigma = 0.0209$ N/m and density $\rho = 0.965\,10^3$ kg/m^3. The forcing frequency $f_0 = 80$ Hz is fixed. Similar results were obtained with an oil of viscosity $\mu_2 = 50 \cdot 10^{-3}$ Pa · s and $f_0 = 50$ Hz. A necessary requirement is to be able to generate on the fluid surface a set of identical droplets of a size of the order of 0.7 mm. The drops are created by dipping a conical pin in the bath, then pulling it out swiftly at a well-defined velocity. Fast camera recordings show the formation of a liquid bridge between the pin and the bath. This bridge pinches off near the two menisci (and only there) and the liquid thread retracts to form a single droplet. The size of this droplet is determined by the depth at which the cone had been plunged into the liquid. By choosing this depth, monodisperse droplets with diameters in the range $0.1 < D < 1.5$ mm could be reproducibly generated.

Binding of two droplets. – At low forcing amplitude, the drop motion is composed of a series of identical jumps and each drop emits a travelling wave of frequency f_0. This wave has a small amplitude and its wavelength λ_0 corresponds to that predicted by the dispersion relation:

$$\omega^2 = \left[gk + (\sigma/\rho)k^3\right] \tanh(kh). \qquad (1)$$

The non-local interaction between drops is due to these surface waves. When two drops interact they come to bounce at a fixed distance from each other. Several distances of equilibrium are possible. The measured values of d_m^0 form a discrete set linearly related to the wavelength λ_0 of the surface waves at the forcing frequency ($\lambda_0 = 2.85$ mm in our experimental conditions). They can all be

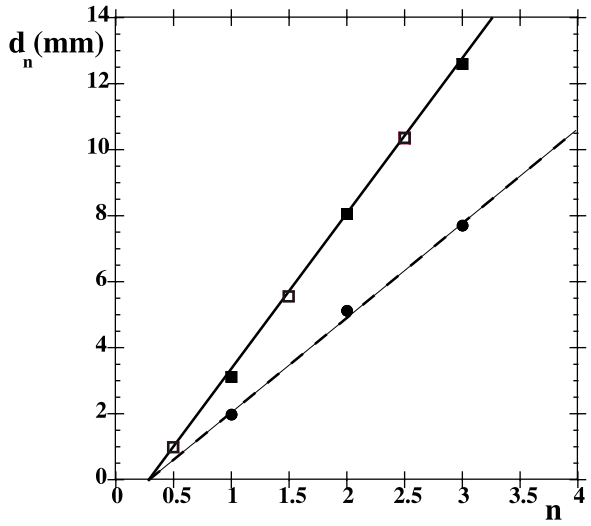

Fig. 3: The possible distances of binding of two drops as a function of the order of the bond linking them. Black dots: drops bouncing at the forcing frequency. With $f_0 = 80$ Hz and $\lambda_0 = 2.85$ mm, we find $d_1^0 = 1.98$ mm, $d_2^0 = 5.12$ mm, $d_3^0 = 7.70$ mm. The squares show the distances of binding of two drops having a period-doubled bouncing. With Faraday frequency $f_F = 40$ Hz and $\lambda_F = 4.73$ mm we find: black squares: $d_1^F = 3,12$ mm, $d_2^F = 8.06$ mm, $d_3^F = 12.6$ mm. Open squares: drops bouncing with opposite phases. $d_{1/2}^F = 0.99$ mm, $d_{3/2}^F = 5.56$ mm, $d_{5/2}^F = 10.35$ mm.

written

$$d_m^0 = (m - \epsilon_0)\lambda_0, \tag{2}$$

where m, the order of the bound state, is one of the integers $m = 1, 2, 3, \ldots$ (see fig. 3). These discrete values are all close to a multiple of the wavelength of the surface wave but shifted by an offset which is the same for all bound states [3]. The offset $\epsilon_0\lambda_0$ is due to the finite duration τ of the contact with the interface. The wave is emitted at lift-off and affects the other drop at its landing so that the shift can be estimated to be approximately half the distance travelled by the wave during the time τ of the collision. Writing $\epsilon_0\lambda_0 = V_\phi\tau/2$ (with $V_\phi = 189$ mm/s the phase velocity of the wave) gives $\tau = 810^{-3}$ s which is of the order of the collision duration.

When the forcing amplitude γ_v/g is increased, there is a threshold (see the phase diagram in [3]) at which the vertical motion of a droplet becomes sub-harmonically modulated. A droplet then undergoes a succession of large and small bounces. Above this threshold, two states are possible as the droplet can have its larger bounce either during one period of the forcing motion or during the next. This is a breaking of symmetry and we will call these distinct states (+) or (−), respectively. The droplet now emits a superposition of waves of frequencies f_0 and $f_0/2$. On a vibrating bath, the waves, parametrically forced by the Faraday instability, have a sub-harmonic frequency $f_F = f_0/2$. Though our experiment is done below the threshold of this instability, these waves are less damped than the others and become dominant when γ_m/g is increased. For this reason, when two drops bind, their possible distances of equilibrium are now related to the wavelength λ_F at the frequency f_F ($\lambda_F = 4.73$ mm in our experimental conditions). They can be written

$$d_n^F = (n - \epsilon_F)\lambda_F, \tag{3}$$

where $\epsilon_F = 0.2$. In contrast with the result given by relation (2), the possible values of n in (3) form two subsets corresponding to drops bouncing in phase or with opposite phases. When in phase, their possible distances $d_n^F(+,+)$ or $d_n^F(-,-)$ satisfy relation (3), with n being one of the successive integers $n = 1, 2, 3, \ldots$. When of opposite phases, $d_n^F(+,-)$ or $d_n^F(-,+)$ satisfy relation (3) with $n = 1/2, 3/2, 5/2 \ldots$. The values of these distances are plotted as a function of n in fig. 3. In the stable arrangements each drop is at such a position that its collisions with the interface do not lead to a horizontal displacement. In the following, we call n the order of the bond.

Aggregates. – We can now examine the situations in which many droplets are present simultaneously on the bath. Below the threshold of period doubling they are observed to bind to each other at one of the d_m^0 and form clusters with various 2D crystalline structures. Lattices with either square or rectangular structures are weakly unstable and relax slowly to the triangular patterns, the most stable having hexagonal symmetry. Structures of the type shown in fig. 1 can thus form spontaneously. When the forcing is increased the transition to a period-doubled bouncing generates an interesting phase transition. At the period doubling threshold, each elements of the periodic lattice undergoes a transition to either of two possible states (+) and (−). Because of the interaction of the droplets, this is a collective process, dominated by frustration effects, which leads continuously to new states with droplets having the two different phases. When the initial crystal is large, this is a solid-state transition. The crystalline structure is destroyed and replaced by a disordered structure which, however, include small organized regions. In the transition of clusters of medium size (twenty droplets), patterns with various polygons formed spontaneously.

In the period-doubled regime the structures obtained with a large number of droplets are generally complex but satisfy general principles of organisation. The observed clusters have two sub-lattices formed by the droplets of each phase. At one forcing period, the droplets of the sub-lattice (+) collide with the interface in troughs of the wave pattern. The swells then become troughs where the droplets (−) fall at the next forcing period. Both sub-lattices contribute to sustain a common, coherent wave field in which all droplets are stable. Of particular interest for the construction of periodic structures are the polygonal rings organized around a droplet of a given phase. These rings can be understood by considering the waves produced by a central drop.

Table 1: A list of some of the possible polygonal assemblies in our experimental conditions. The shape of the elementary triangle is determined by the order of the sides AB, AC and BC, It leads to isosceles (or equilateral) triangles where $\theta_n^{n'}$ is the angle at the main vertex.

AB, AC n	BC n'	$\theta_n^{n'}$	$N_n^{n'} = 360°/\theta_n^{n'}$ polygonal rosettes	Could contribute in Archimedean lattices
1	1	60°	$N_1^1 = 6$	(3^6)
3/2	1	32.6°	$N_{3/2}^1 = 11$	$(3, 12^2)$
3/2	2	92.8°	$N_{3/2}^2 = 3.87 \simeq 4$	$(4^4), (3^2, 4, 3, 4), (3, 4, 6, 4)$
2	2	60°	$N_2^2 = 6$	$(3^6), (3, 6, 3, 6), (3^4, 6), (6^3)$
5/2	2	45.4°	$N_{5/2}^2 = 7.92 \simeq 8$	$(4, 8^2)$

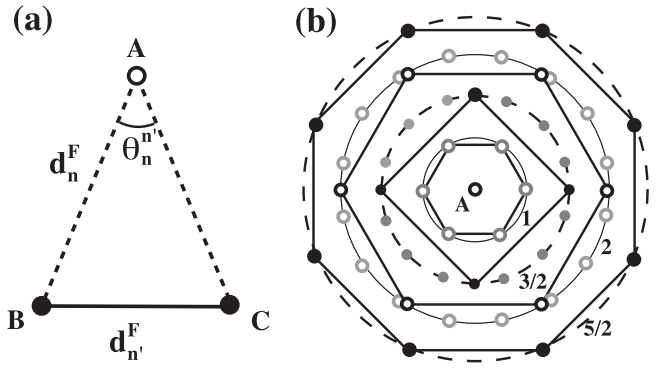

Fig. 4: (a) Sketch of the possible bound states of three drops in the period-doubled state. (b) The possible polygonal rosettes around a drop A of phase (−). The circle of radius d_1^F is saturated when occupied by $N_1^F = 6$ droplets (−) at a distance d_1^F from each other. The next circle $d_{3/2}^F$ can be saturated either by $N_{3/2}^1 = 11$ droplets (+) bound at a distance $d_{3/2}^F$ or by $N_{3/2}^2 = 4$ droplets (+) bound at a distance d_2^F. With the same principle the circle of radius $d_{5/2}^F$ can accommodate $n_{5/2}^2 = 8$ droplets (+) bound at a distance d_2^F.

Let us consider a droplet of phase (−) located at the point A generating concentric circular waves (fig. 4). The circular troughs of this wave are the loci where other droplets can be at rest. Droplets of phase (−) will be stable in the troughs of radius $d_n^F(-,-)$, droplets of phase (+) in those of radii $d_n^F(-,+)$. If several droplets of the same phase are located in the same circular trough they bind to each other at a distance which is necessarily one of the $d_n^F(+,+)$. They thus form, with A, a polygonal rosette formed of juxtaposed isosceles triangles. In the self-organization of small clusters, these rosettes appear spontaneously. We can consider the triangle formed by A and two neighbouring drops in B and C (fig. 4(a)). Calling n the order of the bounds to A and n' the order of the bound of B with C, the angle of the vertex in A will satisfy: $\sin(\theta_n^{n'}/2) = d_{n'}^F/2d_n^F$. The triangle ABC is equilateral if $n' = n$, isosceles in the other cases. The possible values of $\theta_n^{n'}$, deduced from the values of d_n^F are given in table 1. For pairs of values of n and n', the value of $\theta_n^{n'}$ gives the limit number $N_n^{n'} = 2\pi/\theta_n^{n'}$ of droplets which saturates a given trough and the possible symmetry of the rosette.

As shown in table 1 and fig. 4(b), it is possible to assemble around A rings of droplets of the same phase, bound at d_n^F forming polygonal rosettes (fig. 3(b)). These rosettes are observed to form spontaneously in small aggregates and we wondered if they could be the building blocks of Archimedean tilings. We can now revisit the structures shown in fig. 2 and examine them by the types of polygons they contain to see if they can be obtained in our system.

The assembly in equilateral triangles is the simplest and most commonly observed clustering mode of droplets the same phase. The resulting triangular organisation (3^6), being the most dense, self organizes spontaneously as shown in fig. 1. The next organization (4^4) is formed of squares. It is found to be unstable when constituted of droplets of only one phase but it can be obtained by the assembly of identical square rosettes of side d_2^F where four drops of one phase surround at a distance $d_{3/2}^F$ a droplet of the other phase. The assembly of such rosettes gives a "square centred" structure shown in fig. 5(b). It is formed of two identical sub-lattices of droplets (+) and (−) respectively, each formed of squares of side d_2^F and shifted diagonally by $d_{3/2}^F$. Crystallites having this organisation were observed in the spontaneous aggregation of small clusters. However to obtain large crystals we had to resort to an artificial assembly method. After forming a nucleus we keep adding droplets, each one being displaced and released near its predicted position of equilibrium to which it then moves spontaneously. Once formed it is a very stable structure. Two tilings, the snub square $(3^2, 4, 3, 4)$ (fig. 2(d)), and the elongated triangular tiling $(3^3, 4^2)$ (fig. 2(e)) are formed of triangles and squares. They can be obtained by the assembly of the same square rosettes separated by equilateral triangles of sides d_2^F. The resulting clusters are shown in fig. 5(d) and (e), respectively. In the snub square the stabilizing sub-lattice is square. In the elongated mode the stabilizing sub-lattice is parallelogramic. Both are very stable arrangements. It is worth mentioning that in these structures the density of droplets of each phase is not the same.

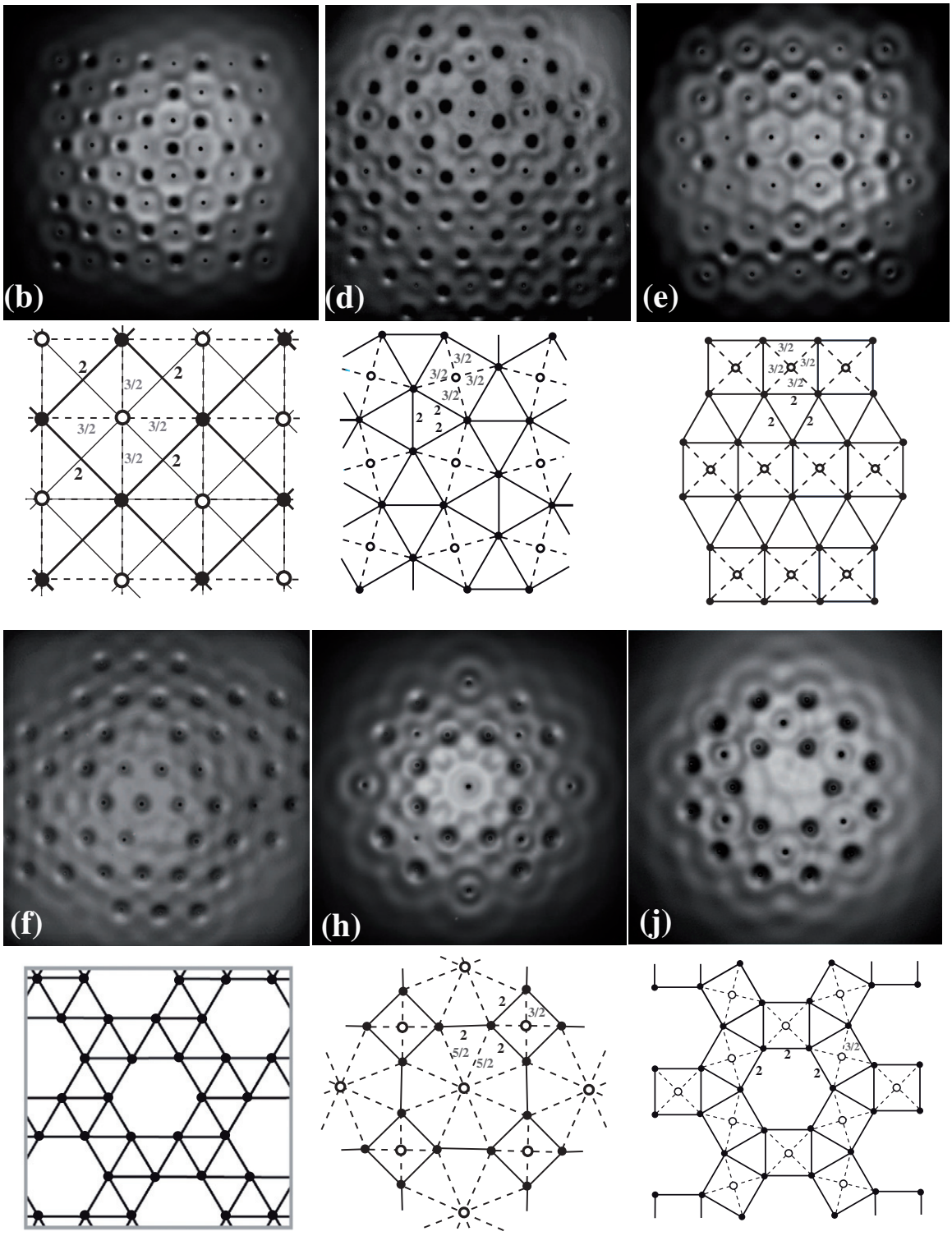

Fig. 5: The observed 2D periodic organizations of droplets of two different phases and their interpretation, labeled by reference to their order in fig. 2. A sketch of the relation between the two sub lattices is given below each photograph. (b) Two identical square sub-lattices (4^4) of side d_2^F. Each square is stabilized by a central drop of the other phase located at a distance $d_{3/2}^F$ of its vertices. Stoichiometry: $R = N_-/N_+ = 1$. See `squares.mov`. (d) A snub square $(3^2, 4, 3, 4)$ associated to a square sublattice. The polygons of the Archimedian sublattice have a side d_2^F. The other sub-lattice stabilizes the small squares, being located at a distance $d_{3/2}^F$ of their vertices. $R = 1/2$. (e) An elongated triangular lattice $(3^3, 4^2)$ associated to a monoclinic sublattice. $R = 1/2$. (f) A snub hexagonal $(6, 3^4)$ pattern obtained by creating voids in a triangular structure. (h) A truncated square $(4, 8^2)$ associated to a square (4^4) sublattice. The polygons have a side d_2^F. The octagons are stabilized by central droplet $(-)$ located at a distance $d_{5/2}^F$. $R = 1/2$. See `Truncated_square.mov`. (j) A small rhombitrihexagonal $(3, 4, 6, 4)$ lattice associated to a trihexagonal $(3, 6, 3, 6)$ sublattice. $R = 3/7$.

Choosing $(-)$ to be the stabilizing sub-lattice with the smaller density of droplets, we define the stoichiometry of the system as $R = N_-/N_+$. In both these lattices $R = 1/2$.

We can now examin the lattices involving either only hexagons (6^3) (fig. 2(c)) or triangles and hexagons: the snub hexagonal tiling $(6, 3^4)$ (fig. 2(f)) and the trihexagonal tiling $(3, 6, 3, 6)$ (fig. 2(g)). They can be seen as lacunary triangular lattices. In order to observe them we assembled triangular crystals and removed droplets so as to generate periodic lacunae. The wave pattern, which is the same as in the triangular lattices, is sustained by a smaller density of droplets, so that these structures are less stable. The hexagonal and trihexagonal structures are unstable and their hexagonal voids tend to collapse. The snub hexagonal $(6, 3^4)$ can be obtained and is shown in fig. 5(f). The structure $(3, 4, 6, 4)$ shown in fig. 2(j) involves triangles, squares and hexagons. Using the same principles we were able to assemble a crystal which associates a small rhombi-trihexagonal $(3, 4, 6, 4)$ sub-lattice with a stabilizing trihexagonal $(3, 6, 3, 6)$ sub lattice. It is stable and shown in fig. 5(j). The stoichiometry is $R = 3/7$. As shown in table 1 and fig. 2(f) octagonal rosette of side d_2^F can form at a distance $d_{5/2}^F$ around a droplet of the other phase. Square and octagonal rosettes can be assembled into a truncated square $(4, 8^2)$ sub-lattice associated with a square sub-lattice (fig. 5(g)). The stoichiometry is $R = 1/2$. As shown on table 1 approximations of dodecagonal rosettes can be obtained with $n = 3/2$ and $n = 1$. We were able to assemble small aggregates having the predicted organisation but they turned out to be unstable. As a result we were not able to obtain the truncated hexagonal tiling $(3, 12, 12)$, nor the great rhombi-trihexagonal $(4, 6, 12)$ tiling.

Conclusion. – The large-scale symmetry in this system results from two types of ordering. At a small scale, a limited number of droplets aggregate. Several stable structures are observed, forming small ordered domains: the rosettes. In these domains the droplets are located at the antinodes of the waves, but not necessarily at all of them. As in the Faraday instability (and unlike in a crystal) a resonant wave-field exists in each of these regions of finite spatial extent. It is sustained by the local forcing of the droplets. These rosettes can be assembled into large structures with the possibility of coexistence of various regular convex polygons. The periodicity results from the periodic arrangement of these domains. Since the wave amplitude decreases away from the sources, the interaction with second neighbours is weak so that the wave structure does not have necessarily the periodicity of a global standing wave. As in a crystal (but differing from Faraday waves), long-range order results here from the clustering of finite size domains. We used this possibility to obtain Archimedean lattices.

These lattices have been obtained by artificial assembly in the period-doubled regime. A different approach would be to assemble a crystal in the simple-bouncing regime and observe result of the phase transition which occurs when the bouncing becomes sub-harmonic. As usual solid-state phase transitions it induces a complex reorganisation into various crystallites with no long-range order, reminiscent of complex structures obtained in the Faraday patterns forced with two frequencies [14]. Numerical simulations, using models in which the multivalued distances of binding are taken into account, could be useful to investigate it. In conclusion, several aspects of this system should trigger further developments. The above-described periodic structures are, once formed, very stable. When the forcing amplitude is strongly increased, they are affected by collective oscillations where all droplets vibrate around their equilibrium positions. These oscillatory modes, which are similar to the phonons of a solid crystal will be described elsewhere. Finally, when the period doubling is complete, the droplets become walkers [3]. The crystal is usually destroyed but small crystallites sometimes undergo a transition to global rotation.

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