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HARTMUT LÖWEN

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Perspective

Active colloidal molecules

Hartmut Löwen

Institut für Theoretische Physik II: Weiche Materie, Heinrich-Heine-Universität Düsseldorf
D-40225 Düsseldorf, Germany

Abstract – Like ordinary molecules are composed of atoms, colloidal molecules consist of several species of colloidal particles tightly bound together. If one of these components is self-propelled or swimming, novel "active colloidal molecules" emerge. Active colloidal molecules exist on various levels such as "homonuclear", "heteronuclear" and "polymeric" and possess a dynamical function moving as propellers, spinners or rotors. Self-assembly of such active complexes has been studied a lot recently and this perspective article summarizes recent progress and gives an outlook to future developments in the rapidly expanding field of active colloidal molecules.

Introduction. – By definition, a *molecule* is a group of two or more atoms held together by chemical bonds. It is a cornerstone of modern chemistry and physics that matter is composed of such molecules which are themselves groups of atoms from the periodic table of elements. The periodic table of elements can therefore be viewed as a "toolkit" to construct and compose molecules and matter in general. There are two different types of molecules: On the one hand, molecules can be composed of identical atoms referred to as *homonuclear* molecules. An example is the oxygen molecule O_2 or the ozone molecule O_3 composed of oxygen atoms O. On the other hand, when molecules are composed of atoms of different (*e.g.*, two) species they are called *heteronuclear*, an example of which is the water molecule H_2O or an organic hydrocarbon chain C_nH_{2n+2} serving as a standard example for a *polymeric* macromolecule, see fig. 1 for an illustration. The typical length scale for the molecular size amounts to a few ångströms or nanometers. Chemical bonding is typically discussed for the case of isolated molecules, *i.e.*, in the gas phase, where the surrounding medium is the vacuum. At room temperature such a molecular gas can often be described as a classical particle system such that temperature excites internal translational, vibrational and rotational modes of the molecules.

The composition principle realized on the molecular scale can be copied on a larger mesoscopic or "colloidal" length scale between a few nanometers and microns. In fact, these colloidal particles can be brought together by attractive effective interaction forces to form a *colloidal molecule*. This was demonstrated at the beginning of this century by using various kinds of colloids including patchy particles with designed attraction zones which form a variety of structures and shapes similar to ordinary molecules [1–9]. Like their atomistic counterparts, the colloidal molecules can be classified into *homonuclear*, *heteronuclear* and *polymeric*, see again fig. 1. As an example of "homonuclear" and "heteronuclear" colloidal molecules, trimers composed of identical spheres ("colloidal ozone molecule") and composed of different spheres ("colloidal water molecule") can be synthesized; see ref. [8] for visualization of such colloidal molecules. Moreover, there are "colloidal polymers" which are chains of linked colloidal monomer representing realizations of macromolecules in the colloidal domain. These system have also been synthesized by various means [10–13]. Mesoscopic colloids offer an important advantage over molecular systems as the individual constituents can be designed at wish [14]. Hence a wide range of colloidal molecules with desired structural and functionalized properties emerges [15], which was an important research line in the last decades. As an important difference to ordinary molecules, colloidal molecules are typically suspended in a molecular carrier fluid indicated as a blue background in fig. 1 which exposes them to thermal fluctuations induced from the solvent leading to their Brownian motion.

The key topic of this perspective article is another recent important step ahead towards *active colloidal molecules*. Active (*i.e.*, motile) colloidal molecules are composed of "active" or "self-propelled" colloids. The latter are
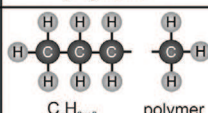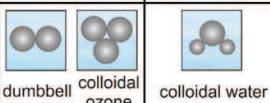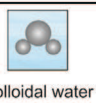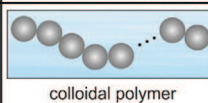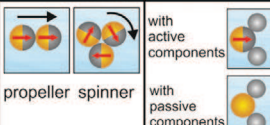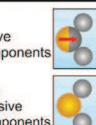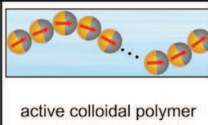
Fig. 1: (Colour online) Classification of ordinary and colloidal molecules (schematic). First row: ordinary homonuclear molecules such as oxygen O_2 and ozone O_3, heteronuclear molecules such as H_2O, and polymeric molecules such as a hydrocarbon chain C_nH_{2n+2}. Second row: colloidal molecules composed of colloidal particles (grey spheres) in a solvent (blue background) including homonuclear dimers and trimers and heteronuclear trimers with two species as well as colloidal polymers. Third row: active colloidal molecules such as homonuclear dimers (translational "propeller") and trimers (rotational "spinner"). An active entity is indicated by a Janus particles with the red arrow indicating its self-propulsion direction. Heteronuclear active colloidal molecules can be divided into two subclasses: i) composed of mixtures of active and passive building blocks, ii) composed of passive building blocks which become active when they meet. Active polymeric colloidal molecules consist of self-propelled monomers.

colloidal particles equipped with an internal motor for their own self-propulsion or, since the particles are embedded in a liquid, for "swimming". Therefore, these particles are sometimes called (synthetic) *microswimmers*. By now there are various means to allow colloidal particles to move on their own, by catalytic reactions, laser-induced diffusiophoresis, external field actuation, etc., see [16–18] for recent reviews. This is highly interesting first of all from a basic physics point of view [19–21] as the particles which consume energy are intrinsically in nonequilibrium. Second, microswimmers are an interesting unanimous realization of moving miroorganisms often leading to functionalities such as navigation [22] or self-healing [23]. Third, many applications become possible such as targeted drug delivery via self-propelled particles [24].

We define *active colloidal molecules* by a group of colloidal particles with the following conditions: i) At least one particle is active (or particles get activated during molecule formation, see below), ii) all particles are interacting by attractive body forces even in the absence of self-propulsion. The second condition implies that collections of particles bound together by pure hydrodynamic interactions are not termed as "colloidal molecules". This definition is not completely sharp. Previously the attractive bond has been postulated to be permanent (like a spring-like force) [25,26] but we generalize it here a bit towards

other strong bonding attraction energies much larger than the thermal energy k_BT. Typically the bonding force is then larger than or comparable to forces arising from self-propulsion. In this context we define an active molecule not as a single swimmer but as an assembly of many swimmers [25]. Many bead models for a single swimmer as the traditional three-bead Najafi-Golestanian swimmer [27] with nonreciprocal dynamics in the relative motion of the beads are in this sense not an active molecule. In this regard, only the special case of heteronuclear molecules with one passive component [28,29] or two passive particles which get active when combined [30] is ambiguous, since such a pair can be simultaneously considered as a "single swimmer" or an "active molecule".

Figure 1 summarizes a possible classification scheme of active colloidal molecules. The simplest homonuclear molecule is composed of two identical self-propelled particles. When combined to a pair by bonding, the dimer propels translationally along the joint orientation. A trimeric homonuclear ring-like aggregate (also shown in fig. 1) leads to a spinning molecule or a rotator. The basic difference between "thermal" colloidal molecules and active colloidal molecules is their dynamic function: active colloidal molecules move on their own. This sets them apart for special dynamic functionalities. Clearly different species can be combined in a next level of complexity leading to heteronuclear molecules [31,32]. In the binary case, two situations can be distinguished: either there is a combination of active and passive components (*e.g.*, active colloids are mixed with ordinary passive colloids) or there is an even more extraordinary case which has no direct analog to ordinary colloidal molecules: two components which are passive on their own become active when exposed and bound to each other thus achieving a dynamical function only when combined [33,34]. We shall discuss experimental realizations of these remarkable molecules in the following [35–37]. Finally, polymeric active colloidal molecules are conceivable as well; these could be long colloidal chains with active monomers leading to an active colloidal polymer or an "active macromolecule" [38].

At this point it is important to distinguish between active colloidal molecules, for which we assume attractive bonding between the different constituents, and purely motility-induced phase separation which nucleates at clusters which occur already for purely repulsive systems [39]. We do not discuss these fragile clusters here in depth since this motility-induced clustering is by now well-established and has been reviewed already [40,41]. Instead we focus on "active molecules" where a real bonding energy (or effective attractive force) exists between the "atoms". However, a strong attractive phoretic attraction between neighboring particles in the experimental samples [42,43] will bring dynamical clusters to the case considered here.

This perspective article is organized as follows: we first discuss a simple minimal model for an active colloidal molecule to illustrate its stability and dynamics. We then review more sophisticated modelling and theoretical

predictions obtained mainly by computer simulations of microswimmer clusters. Then we turn to experimental realizations which have been proposed recently. Finally, conclusions and perspectives of where this rapidly expanding field is developing are provided and some challenges ahead are discussed.

Simplest model of an active colloidal molecule. – Figure 2(a) represents the simplest situation of an active homonuclear colloidal molecule, namely two active particles bound via a spring-like (harmonic) potential, sometimes called an active dumbbell [44]. In two spatial dimensions, the configuration of the two self-propelled particles are described by their center-of-mass positions $\vec{r}_1$ and $\vec{r}_2$ and their orientational unit vectors $\hat{u}_1 = (\cos\phi_1, \sin\phi_1)$ and $\hat{u}_2 = (\cos\phi_2, \sin\phi_2)$ with ϕ_i $(i = 1, 2)$ denoting the angle of the particle orientation with respect to the x-axis. These unit vectors provide the actual directions of the self-propulsion which is an inner degree of freedom attached to the particles. Since there is a surrounding fluid and the particles are micron-sized, we are typically in the low-Reynolds-number regime and the simplest description is a completely overdamped dynamics for the translational and orientational motion $(i = 1, 2)$ neglecting any hydrodynamic interactions mediated by the flow field of the solvent. Then the equations of motion for the translational and orientational degrees of freedom read as follows:

$$\gamma\dot{\vec{r}}_i = \gamma v \hat{u}_i - \frac{\partial}{\partial \vec{r}_i} V(|\vec{r}_1 - \vec{r}_2|) + \vec{f}_i(t) \tag{1}$$

and

$$\gamma_R \dot{\phi}_i = g_i(t). \tag{2}$$

Equation (1) can be viewed as a force balance. The term $\gamma\dot{\vec{r}}_i$ on the left-hand side is the Stokes drag force with γ denoting the Stokes drag translational friction coefficient. The term $\gamma v \hat{u}_i$ represents the self-propulsion which couples translation and rotation in a nontrivial way where v is the imposed self-propulsion speed. The spring interaction force $-\frac{\partial}{\partial \vec{r}_i} V(|\vec{r}_1 - \vec{r}_2|)$ is derived from a harmonic potential

$$V(r) = \frac{k}{2}(r - \ell)^2 \tag{3}$$

of a spring with finite rest length ℓ and spring constant k. Clearly the spring interaction force is reciprocal, *i.e.*, *actio* is minus *reactio*, such that Newton's third law is fulfilled. Finally, $\vec{f}_i(t)$ are Gaussian random forces ("noise") stemming from the solvent kicks with zero mean and a variance between two Cartesian components α and β of $\langle f_i^\alpha(t) f_j^\beta(t')\rangle = 2k_B T \gamma \delta_{\alpha\beta}\delta_{ij}\delta(t - t')$, where T denotes some effective noise strength or effective temperature. Note that T does not necessarily correspond to the real temperature as we are far from equilibrium. Likewise eq. (2) represents a torque balance between Stokes drag torque $\gamma_R\dot{\phi}_i$ on the left-hand side and a random Gaussian torque $g_i(t)$ with zero mean and variance
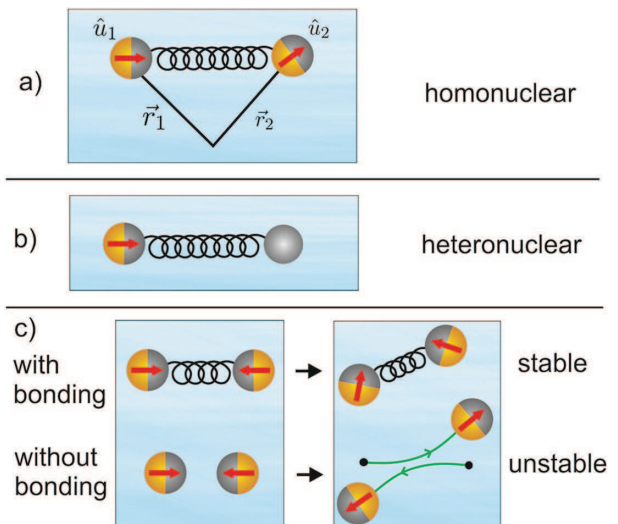
Fig. 2: (Colour online) Simple models for an active colloidal molecule. (a) Homonuclear active colloidal molecule composed of two identical self-propelled particles at central positions $\vec{r}_1$ and $\vec{r}_2$ and connected by an effective spring as a "chemical bond". The self-propulsion direction (or internal propulsion force) is indicated as a red arrow along the unit vectors $\hat{u}_1$ and $\hat{u}_2$. (b) Heteronuclear active colloidal molecule composed of a self-propelled particle and a passive particle. (c) Homonuclear active colloidal molecule with two opposing self-propulsion forces leading to an overall stable cluster due to the spring. In the absence of any spring, the pair is long-lived on a time scale set by the inverse rotational diffusion constant but unstable for long time unless further neighbouring particles accumulate on the pair. The latter situation represents a simple case for motility-induced clustering in the absence of bonding.

$\langle g_i(t) g_j(t')\rangle = 2k_B T \gamma_R \delta_{ij}\delta(t - t')$. Here γ_R is the rotational friction coefficient.

It is instructive to consider a configuration for a homonuclear pair where the two particles have opposing propulsion directions as shown in fig. 2(c). In fact, if the two self-propulsion forces are exactly opposing and are not too large (*i.e.*, $v > k\ell/\gamma$), there is a distance where they are exactly compensated by the repulsive spring forces. Therefore, in the absence of noise, a pair will not move in this special configuration. This static configuration can be characterized by a six-component multivector $(\vec{r}_1^*, \vec{r}_2^*, \phi_1^* = \pi, \phi_2^* = 0)$ summarizing the positions and orientations of the two particles. Expanding the equations of motion around this static configuration to linear order in the six difference variables $X = (\vec{r}_1(t) - \vec{r}_1^*, \vec{r}_2(t) - \vec{r}_2^*, \phi_1 - \phi_1^*, \phi_2 - \phi_2^*)$ yields a matrix equation of the form

$$\dot{X} = \bar{\bar{A}} \cdot X, \tag{4}$$

where $\bar{\bar{A}}$ is a 6×6 "dynamical" matrix which contains real and constant matrix elements. The corresponding eigenmodes around the static reference point $X = 0$ are obtained by plugging an ansatz into eq. (4) of the form

$$X(t) = X_\lambda(0)\exp(\lambda t), \tag{5}$$

where the real part of λ is the growth rate and $X_\lambda(0)$ the associated eigenvector. If a solution with a positive real part of λ is possible, the configuration is unstable with respect to the noise, while a negative λ means a damped eigenmode that is stable against the noise. For the intermediate vanishing case, $\lambda = 0$, the situation is marginal or neutral; this often reflects a continuous symmetry in the system which leads to a zero eigenmode.

Let us first discuss the simplest reference case where the spring is applied and has a finite extension ($\ell > 0$, $k > 0$) but the self-propulsion force is vanishing ($v = 0$). In this case all forces are reciprocal, the 6×6 matrix $\bar{\bar{A}}$ is symmetric and can be diagonalized. One eigenvalue is negative and the remaining five eigenvalues are zero. The latter correspond to five different symmetry operations such as: i) global translation in the x-direction, ii) global translation in the y-direction, iii) global rotation of the whole system, iv) individual internal rotation of the left particle, v) individual internal rotation of the right particle. The eigenmode belonging to the negative eigenvalue corresponds to a radial distortion of the particle pair, this is kept stable against the noise via the attractive bonding.

If the self-propulsion is finite ($v > 0$), the matrix is not any longer symmetric, this is a consequence of the fact that self-propulsion leads to nonreciprocal interactions [33,34,45]. Nevertheless there is still one negative eigenvalue which again corresponds to the radial distortion kept stable via the harmonic bonding. But there are not any longer 5 clean zero eigenvalues. Remember that not any real unsymmetric matrix can be diagonalized (even not in the complex), it can only be brought to the Jordan normal form in a suitable basis. This is a basic difference to passive colloidal molecules where the dynamical matrix is symmetric and diagonalizable. In particular, in the special case of vanishing spring constant k (see fig. 2(c) again), there are neither stable nor unstable modes but neutral or marginal ones. This configuration is not stable: following weak noise the particles will scatter around each other and move away from each other, see the possible trajectories for self-propelled particles in fig. 2(c). This set-up corresponds to pure motility-induced clustering without any bonding pointing to the finite life of any cluster for self-propelled particles with repulsive interactions, see also possible trajectories for self-propelled particles in fig. 2(c).

A heteronuclear situation shown in fig. 2(b) can be described by similar overdamped equations of motion (1), (2) [26] except for the fact that rotational motion is irrelevant for a passive sphere. The stability analysis is similar in spirit than for the homonuclear pair.

The simple example shows two basic facts: i) attractive bonding is needed for stable clusters ii) the motion of a pair or a general molecule is more complicated than that of the individual self-propelled particles.

Theory and simulation of active colloidal molecules. – Let us now discuss recent models for active colloidal molecules which were explored by theory and computer simulations. Experimental realizations are subsequently mentioned in the next section.

Spring-like interaction forces. Harmonic springs (see eq. (3)) constitute the simplest models for bonding. In particular these forces are obtained when expanding the equations of motion around a stable configuration, see again the discussion in the previous section. Heteronuclear active colloidal molecules where one active particle is linked to passive spheres via springs were proposed in ref. [26], see also [46]. The resulting active colloidal molecules can alternatively be viewed as a deformable swimmer. The importance of deformability was emphasized when the swimmer is exposed to external vortical flow fields [26,47]. It turns out that the degree of deformability decides whether the swimmer is getting drowned in a swirl or not [26].

If several active particles are connected by springs along one dimension we arrive at the polymeric case of an active polymer chain, see again fig. 1. The first model for active polymers was proposed by Kaiser *et al.* [48]. It is the natural extension of the harmonic model discussed for two particles in the previous section to a long chain. In this sense it corresponds to the active extension of the traditional Rouse model [48–50]. As a result, it was found [48] that the polymer swelling is significantly affected by activity. Subsequently, other models for an active polymers were proposed and discussed such as active worms [51] and active filaments [52,53] and polymers with nonthermal active noise [54,55] mimicking the internal activity of the monomers. For more details we refer to the recent review in ref. [56].

Finally, a full two-dimensional network of springs connecting active particles to a solid sheet has been proposed and analyzed by Ferrante *et al.* [57] revealing new dynamical modes. This can be considered as a limit of a flat active colloidal macromolecule, analogous to an active molecular graphene sheet.

Active colloidal molecules governed by dipolar interactions. The interaction between two dipoles depends on their relative orientation and position, and it can be both attractive and repulsive. The attractive case leads to a tendency of chaining in dipolar suspensions. Recently clusters of active colloidal particles kept together by (*e.g.*, magnetic) dipolar attractions have been realized [58]. These are an ideal set-up for magnetic active molecules. The classical ground state minimizing the total interaction energy of N hard-sphere dipoles with a central permanent dipole moment is a linear chain for $2 \leq N \leq 3$ and a ring for $4 \leq N \leq 13$ [59]. Adding self-propulsion to these molecules induces novel effects. These have first been explored in the absence of hydrodynamic interactions [60]. As a result, a linear chain propagates and a ring rotates but they are stable for any self-propulsion speed v. Starting with an initial metastable Y-like configuration results in seven different steady states including a *fission* of the initial Y-junction [60]. This demonstrates

that self-propulsion has a subtle effect on molecules formation and structure.

Subsequently the effects of hydrodynamic interactions mediated by the solvent on the motion of dipolar clusters were explored. First of all, a straight trimer of hard-sphere dipoles with permanent harmonic bonds was considered in ref. [25]. With increasing self-propulsion, an oscillatory instability was observed which is accompanied by a corkscrew-like swimming trajectory of increasing radius. Hydrodynamic interactions in active magnetic clusters were studied as well by Guzmán-Lastra *et al.* [61]. It was shown that fission and fusion processes of colliding clusters may occur resulting from a competition between dipolar and magnetic interactions. For instance, a linear dipolar chain, which would be absolutely stable in the case of neglected hydrodynamic interactions [60], will break into segments if the particles swim as "pushers" but remain stable if they are "pullers" [61].

Attractive chemotaxis. Many microorganisms but also synthetic colloids sense a chemical which is emitted by other particles and adjust their motion towards the positive or negative gradient of the chemical concentration field, a phenomenon which is called *chemotaxis.* It is an old idea of Tsori and de Gennes [62] that particles which are coupled chemotactically are equivalent to a classical Coulomb system [63]. The underlying reason for the "Coulomb analogy" is that the stationary solution for the diffusion equation with a constant ejection source is an orbital around the secreting particle which decays as the inverse distance, formally exactly the same behaviour as for the classical Poisson equation of electrostatics [63]. If in a two-component system, one species is attracted towards the chemical which is secreted by the other species (chemo-attraction), chemotaxis is formally identical to a Coulomb interaction although there is more freedom as the interaction forces are not necessarily reciprocal [34,37,64,65]. This provides an enormous possibility to construct active colloidal molecules with a dynamic function such as movers, rotators and circle swimmers. In the opposite case of chemo-repulsion there is no bonding and hence no colloidal molecules emerge.

In this context, it is important to note that the particle of each species on their own are passive, *i.e.*, they would not move in the absence of particles of the other species. Hence passive building blocks merge and spontaneously acquire activity by merging. This is what is shown in the third row of fig. 1.

Other attractive interactions. Other types of attractive interaction for microswimmer bonding have been proposed. In ref. [66] clusters arise from a lock-and-key shape of particles like triangular shapes with an opening angle of $2\pi/n$ with $n > 1$ an integer number. These clusters are stabilized by additional attractions and may thereby considered to be active colloidal molecules. Cluster bounds via attractive patches were also proposed and studied [67] driven by the general idea to use patchy colloids as active

systems. Also swimmers interacting with the traditional Lennard-Jones pair potential were studied in order to access their nonequilibrium critical behaviour [68]. For strong enough bonding these clusters are pretty stable exhibiting active colloidal molecules.

Finally recently binary passive building blocks were studied which, when merged, get active, such as two individual hemispheres of a split Janus particle. The attraction was modelled close to an experimental realization to be an attractive Casimir force appropriate as an effective interaction between the passive building blocks when immersed in a near-critical solvent [37]. In this model a variety of active colloidal molecules was discovered, too.

Experimental realization of active colloidal molecules. – The wonderful world of active colloidal molecules is not pure fiction but these artificial complexes have meanwhile been largely synthesized and realized. It is instructive to consider the classification as shown in fig. 1 again and draw actual micro-images from recent experiments instead of the schematic images. This is summarized in fig. 3. In fact all the different colloidal molecules shown there have been fabricated and discovered. In what follows we review some basic progress in these experiments.

First of all a harmonic or spring-like bonding can be achieved by attaching DNA bundles onto active colloids as recently demonstrated [13,69]. This was shown for a chain of passive particles which can be activated by an external magnetic field [69]. Moreover a DNA bundle was used as a flagellum driving a single colloidal sphere [70] but it can likewise also be used to bind two active spheres permanently. A dimer composed of two active Janus particles was studied as the simplest *homonuclear* colloidal molecule [71,72]. Homonuclear molecules consisting of two to four metallic rods were found to exhibit interesting structures such as a T-shape in ref. [73], see also fig. 3. Electric-field controlled structures of active molecules including *active colloidal polymers* were proposed in refs. [38,74,75]. Another homonuclear "polymer" consisting of four cubic monomers was found in ref. [76]. *Heteronuclear* assemblies of active and passive colloids were observed for catalytically powered [77,78], light-activated [79,80] and electric-field controlled [81] self-propulsion mechanisms. Finally, a rich self-organization kit for constructing tight colloidal molecules on various levels using a sequential capillary assembly was provided [82].

Second, there is a broad variety by now for *magnetic dipolar swimmers* self-assembled to active molecules. These can be chains of propellers or rotators [83,84] as well as elongated magnetic particle clusters [58] or large colloidal asters [85] energized by an alternating magnetic field [86].

Finally there are several examples for *passive entities which form an active molecule when combined.* The first example is a so-called modular swimmer consisting of an
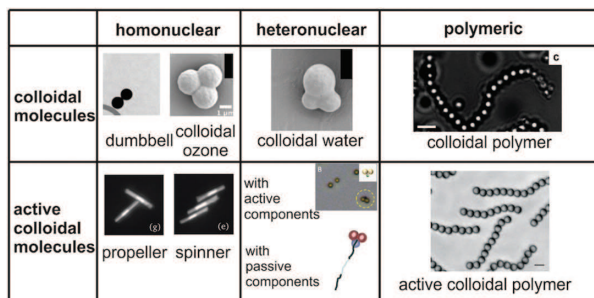
Fig. 3: (Colour online) Same as fig. 1, but now not schematic but with actual realizations of active colloidal molecules. First row: colloidal molecules composed of colloidal particles including homonuclear dimers (image from ref. [87]) and trimers (image from ref. [8]) and heteronuclear trimers (image from ref. [8]) with two species as well as a colloidal polymer (image from ref. [11]). Second row: active colloidal molecules such as homonuclear dimers (translational "propeller") (image from ref. [73]) and trimers (image rotational "spinner") (image from ref. [73]). Heteronuclear active colloidal molecules: i) composed of mixtures of active and passive building blocks (image from ref. [80]), ii) composed of passive building blocks which become active when they meet (image from ref. [37]). Active polymeric colloidal molecules (image from ref. [38]).

ion-exchange resin and passive colloids as proposed by Niu *et al.* [35,36]. The flux of ions towards the resin causes flow which advects passive colloids resulting in a moving composite object which is a colloidal molecule. For a pure suspension of ion exchangers either one-component or two-component (cationic and anionic) exchangers colloidal molecules were also obtained [35,36]. Finally two kind of passive spherical beads interacting via attractive Casimir forces were studied by Schmidt *et al.* [37] which become active under light illumination when merged. This provides an important control of active molecules and their formation via external light.

Conclusions and challenges ahead. – In conclusion we have outlined a novel route towards active complexes as "active colloidal molecules". Like ordinary molecules are composed of atoms, the same idea can be applied to the colloidal mesoscale by creating new active composite aggregates merging single active particles. The self-organization into stable active clusters which possess characteristic dynamical and structural functions has revealed new nonequilibrium physics and is promising for many applications such as functionalized nanomachines.

In the field of active colloidal molecules, the following challenges are lying ahead:

i) Once single molecules are established, a possible next step is to mix them to obtain more complex colloidal structures. These can be based on a lock-and-key principle, on other complex patchy bonding or on hydrodynamic lubrication [88].

ii) Again, once single molecules are established, the next step is to go to finite concentration and condensed

phases. One will expect a complex "zoo" of different living structures for strongly interacting systems. Few examples include: motility-induced clustering for active colloidal molecules, active crystals of colloidal molecules which are expected to have a rich dynamics and a complex melting scenario [89–92].

iii) Another next natural step is to move from few-particle molecules to "colloidal macromolecules" with new hierarchial structures on a supramolecular scale. There is an almost unlimited potential to combine new building blocks on any scale to arrive at complex particles which can possess interesting structures. One example are hollow particles which push solvent through the holes as recently realized for active sperm attached to a hollow tube, so-called spermbots [93,94].

iv) So far we have discussed active colloidal molecules in a Newtonian fluid. New physics arises if the background medium is changed towards a complex fluid which can be viscoelastic (see, *e.g.*, [95]), or if the background itself is another kind of soft matter.

v) In general active colloidal molecules will challenge existing theories for nonequilibrium systems such as dynamical density functional theory (see, *e.g.*, [96]) or mode coupling theory (see, *e.g.*, [97,98]). These theoretical approaches need to be generalized or completely newly founded if activity comes into play.

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