

Figure 1a summarizes the light-extraction process from TADF emitters. Excitons at the lowest triplet state (T_1), generated by carrier recombination with a probability of 75%, generally deactivate without emission in conventional fluorescent molecules, whereas in TADF molecules they can convert to the lowest excited singlet state (S_1) by reverse intersystem crossing (RISC) and be extracted as delayed fluorescence. The overall efficiency of this process depends on the fluorescence rate from S_1 (k_F) and the RISC rate (k_{RISC}), which should be fast enough to outcompete alternative non-radiative deactivation paths from S_1 (with rate k_{NR}^S) and T_1 (with rate k_{NR}^T). A large oscillator strength from S_1 (f) and a small energy difference between S_1 and T_1 (ΔE_{ST}) both increase k_F and k_{RISC} (refs 8,9).

As a first step, Aspuru-Guzik and colleagues tested several computational algorithms for estimates of f and ΔE_{ST} against parameters extracted experimentally from known molecules. Comparing data from 63 samples with a variety of quantum calculation (QC) methods (Fig. 1b), they observed that time-dependent density functional theory, a popular method for computing electronic excited states, was able to accurately predict the physical parameters of the known molecules. However, QCs are usually time-consuming; they therefore resorted to a neural network to realize an empirical model that is able to provide more rapid estimations of the molecular parameters with the same accuracy as QC calculations.

In particular, they trained the neural network to minimize the root-mean-square error of the rate of TADF (k_{TADF} , a parameter directly related to f and ΔE_{ST}). Molecules with the highest predicted k_{TADF} values were simulated with the optimized QC method,

and the rate parameters obtained were used to iteratively train the neural network (Fig. 1c) until the k_{TADF} values predicted with both methods were comparable (Fig. 1c(ii)). As a result of this machine-learning process, the predicted physical parameters were shown to match those of the 63 experimentally tested molecules used to validate the QC calculations (Fig. 1c(i)).

The trained network was then used to calculate the statistical distribution of the relationships between f and ΔE_{ST} , as well as k_{TADF} , extracted from 400,000 molecules selected from an initial library of 1.6 million samples (Fig. 1d). Nine hundred of these structures showed values of f and ΔE_{ST} that are comparable to those of recently reported TADF emitters with high electron-photon conversion efficiencies (Fig. 1e). Further screening was performed at this stage through a web-based consultation with a pool of experts, who were asked to rank the selected molecules according to the novelty of their structures and the expected ease of synthesizing them (Fig. 1f). Finally, Aspuru-Guzik and colleagues synthesized four of these novel molecules, and confirmed that the predicted f and ΔE_{ST} values matched well with those from experiment. OLED devices using these emitters showed excellent electron-photon conversion yields (Fig. 1g), thus demonstrating the practical effectiveness of this high-throughput virtual screening process. The total CPU time for the screening of 1.6 million molecules was 13 million core-hours, corresponding to approximately two months when a super computer with 10,000 cores is used.

Recently, density functional theory has been used to predict the performance of materials for a variety of applications¹¹. Major

efforts are obviously focused on the accuracy of these calculations; however, controlling the overall computational costs is also crucial, particularly if the tight schedule of materials development in an industrial environment must be met. Successful demonstrations of the predictive potential of neural network algorithms, such as that reported by Aspuru-Guzik and co-authors, promote high-throughput computational methods as a potentially important tool for industry to minimize the development cost of new materials. In addition, this statistical approach may lead, in some cases, to the identification of relationships among structural and physical parameters that could not be anticipated by known theoretical models. The fundamental insight obtained by large-scale statistical data may thus complement and expand that achieved by the analysis of experimental results (which are by necessity more limited in number), and contribute to improving our understanding of materials science. □

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ACTIVE COLLOIDS

Controlled collective motions

Self-propelled Janus particles with externally regulated anisotropic interactions can be made to swarm, cluster and form slithering chains.

Roberto Di Leonardo

Spherical particles with isotropic interactions can spontaneously organize into a limited set of ordered condensed phases, namely close-packed crystals. As soon as the simplest anisotropy appears — the head–tail asymmetry found in amphiphilic molecules or tubulin dimers for instance — a myriad of self-organized structures can be observed, including vesicles, microtubules and

other complex supramolecular architectures that populate the world of soft and biological matter. When the basic units are self-propelled ‘particles’ (for example swimming bacteria), anisotropic interactions can reorient the direction of motion, and lead instead to self-organized dynamical states such as flocks¹ or vortices². The synthesis of colloidal particles that mimic the anisotropic interactions of

complex molecules or the self-propulsion of living organisms has been at the centre of two of the most exciting lines of research in present soft matter science. The first endeavour has been to design particles with ‘patchy’ anisotropic interactions, which could expand the set of achievable self-organized structures in colloidal systems³. The second direction investigates novel active materials

that are based on self-propelled colloids, which use a variety of mechanisms to irreversibly convert environmental energy into directed motion⁴. Although a lot of progress has been made in both areas, there is great potential in the combination of the two fields. Writing in *Nature Materials*, Erik Luijten, Steve Granick and collaborators now demonstrate a fascinating and powerful strategy to produce self-propelled colloids with tunable anisotropic interactions⁵. These particles can switch between different forms of collective motion — flocks, clusters and slithering snakes — by simply tuning a single external parameter.

The basic units are silica beads measuring 3 μm in diameter that have a metallic coating on one hemisphere. The application of an a.c. electric field has two remarkable effects on such Janus colloids when immersed in water. First the electric field acting on the diffuse charge layer it has induced at each particle surface gives rise to unbalanced fluid flows around the two hemispheres. This results in particle self-propulsion, an effect already known as induced-charge electrophoresis⁶. At the same time, the two hemispheres respond to the a.c. electric field by developing two oscillating electric dipoles of different magnitudes and phases. On the dielectric side of the colloids, the dipole coefficient switches from positive to negative when the driving frequency of the electric field is larger than roughly 10 kHz. A similar transition occurs on the metallic particle side, but from a negative to a positive dipole coefficient. As a result the metallic (or dielectric) hemispheres of two particles repel each other, with a force that is dependent on frequency. On the other hand, the metallic side of one particle and the dielectric side of another can display

(depending on the driving frequency) either repulsive or attractive interactions (Fig. 1a).

At low frequencies, interactions are weak because of ionic screening — the particles collide randomly and a disordered, gas-like state is observed (Fig. 1b). When the frequency is increased the repulsion between metallic halves is found to be stronger than the repulsion between dielectric hemispheres. The consequences (in terms of particle behaviour) of these unmatched repulsions depend crucially on the direction of self-propulsion. At low ionic strengths particles move with the metallic face forwards, meaning that the stronger repulsion occurs between the leading hemispheres (the heads), and particle velocities therefore tend to align during collisions. In this situation, beautiful swarms are observed, containing thousands of Janus particles that move coherently like fish in a school or birds in a flock (Fig. 1c). At higher salt concentrations the self-propulsion direction is reversed and particles move with the dielectric side facing forwards. Now, because of the larger repulsion between the metallic ‘tails’, the particles jam head-to-head, forming large, densely packed clusters (Fig. 1d). By further increasing the driving frequency, a strong attraction between the opposite poles of different particles appears. Such a head-to-tail attraction causes active particles to self-assemble into active chains that move around like snakes (Fig. 1e). When two active chains collide they can split and recombine, exchange parts or even form rotating closed loops.

The fact that different anisotropic interactions can lead to a complex dynamical phase space is not completely unexpected. Previous work on this subject (mainly focusing on two-dimensional systems of

self-propelled rod-shaped particles and bacteria) shows that the particle aspect ratio is an important control parameter that triggers the transition between disordered gas-like states and more ordered collective motion (such as flocks or active turbulence)⁷. The aspect ratio, however, is a constitutive property of particles — it has a given value for a specific physical system and cannot be modulated by the action of some external field. In this sense the work by Granick and co-authors introduces a new class of active materials where interactions become another controllable parameter that can be used to program different patterns of collective behaviour into the same physical system. The capability to continuously and dynamically tune interactions will be crucial for studying the onset of collective motion, investigating the boundary between different dynamical phases and answering questions such as: how sharp and well-defined are these transitions? How do new phases nucleate? How do changes in the interactions affect characteristic length scales such as velocity correlations in a flock, or the length of particle chains? A potential drawback in the current approach is that both particle speed and interactions are controlled by the same parameter, namely the frequency of the driving electric field. It will therefore be important to investigate the possibility of expanding the set of control parameters and enabling the tuning of activity and interactions independently. The number density of active colloids is also found to be an important variable in controlling the dynamical behaviour⁸, and a systematic examination of the non-equilibrium phase diagram over the frequency–density plane could reveal new forms of collective motions. □

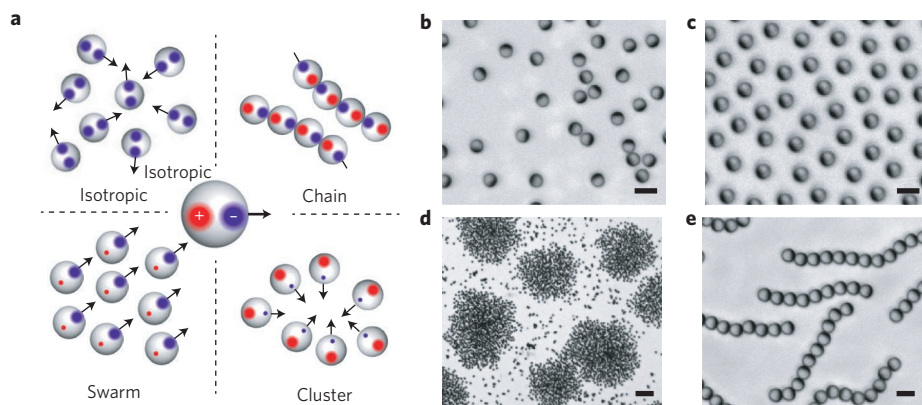


Figure 1 | Realization of various forms of particle collective motion. **a**, The proposed strategy for achieving anisotropic and reconfigurable interactions is based on Janus particles characterized by two effective charges that are displaced from the centre in opposite directions and whose signs and magnitudes are externally tunable. **b**, When the two charges are balanced or weak a disordered gas-like state is observed. **c,d**, Particles experiencing repulsion between the heads self-organize into large flocks (**c**), whereas repulsion between the tails leads to jamming into dense clusters (**d**). **e**, Head-to-tail attraction gives rise to slithering chains. Figure reproduced from ref. 5, Nature Publishing Group.

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