

## ORGANIC LIGHT-EMITTING DIODES

## High-throughput virtual screening

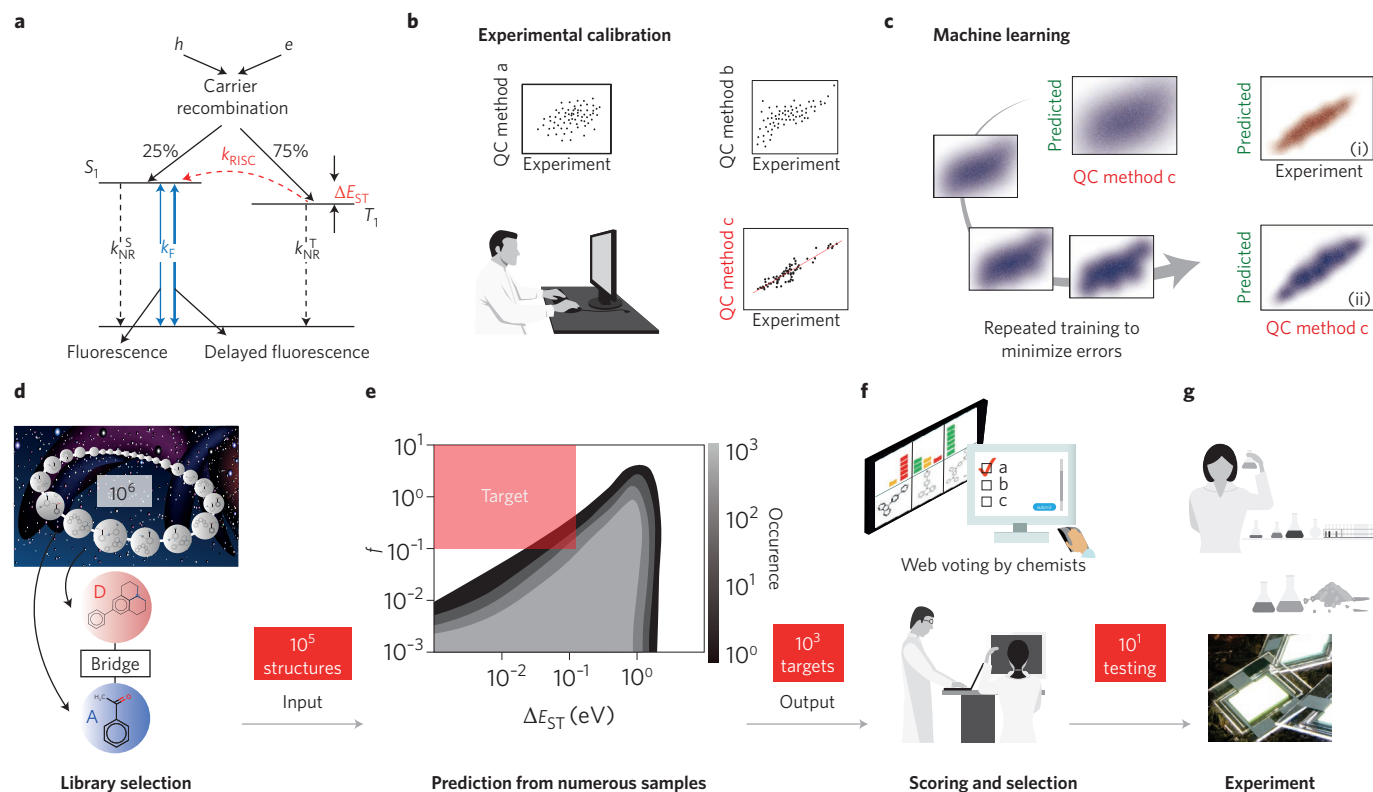
Computer networks, trained with data from delayed-fluorescence materials that have been successfully used in organic light-emitting diodes, facilitate the high-speed prediction of good emitters for display and lighting applications.

Shuzo Hirata and Katsuyuki Shizu

Next year marks the 30th anniversary of the first high-performance demonstration of organic light-emitting diodes (OLEDs). Following that demonstration<sup>1</sup> key performance improvements in the emitting materials used in these devices have marked several milestones on the journey of this technology to its implementation in lighting and displays for mobile devices and televisions. For instance, phosphorescent materials embedding rare-metal complexes that were reported in 1998<sup>2</sup> enabled a 100% electron-photon conversion yield that improved

the internal electroluminescence quantum efficiency obtained with fluorescent emitters by a factor of four<sup>3,4</sup>. In 2012, similar yields were obtained using metal-free aromatic molecules<sup>5</sup> that exhibit thermally activated delayed fluorescence (TADF), a family of materials that may drastically reduce the costs of phosphorescent emitters and improve the operational lifetime of devices emitting in the blue spectral range<sup>6</sup>. Using density functional theory analysis, several design rules have been defined for the synthesis of a few highly-efficient TADF molecules<sup>7–9</sup>. Yet, testing — both computationally and

experimentally — the thousands of potential TADF candidates that may exist in the chemical space is extremely time-consuming. Writing in *Nature Materials*, a collaboration led by Alán Aspuru-Guzik now reports a high-speed protocol that, leveraging high-throughput calculations, machine-learning algorithms and experimental validation, enables fast screening of more than 1.6 million molecules for the identification of TADF molecules that, if used in OLEDs, could lead to external quantum efficiencies that are comparable with state-of-the-art devices<sup>10</sup>.



**Figure 1** | High-throughput virtual screening process leading to the identification of efficient TADF emitters. **a**, Light-generation process in TADF molecules: 25% of the excitons formed by the recombination of electrogenerated charge carriers is extracted as prompt fluorescence from  $S_1$ ; 75% of the excitons form  $T_1$  states that are extracted as delayed fluorescence from  $S_1$  through thermally activated RISC.  $k_{NR}^S$  is the rate constant of internal conversion from  $S_1$ ,  $k_{NR}^T$  is the rate constant of non-radiative deactivation from  $T_1$ . **b**, Pre-screening among QC methods to identify the computational approach that correlates best with the experimental results. **c**, Training of a neural network by machine learning, aiming to optimize the correlation between the physical parameters predicted by the neural network with those calculated with QC methods (ii) and with experimental values (i). **d**, Generation of a library of molecular structures with potential TADF properties that comprise a donor (D) and an acceptor (A) linked by a bridge. **e**, Plot of the statistical distribution of the photophysical parameters derived by the trained neural network and selection of molecules with promising  $f$  and  $\Delta E_{ST}$  values (which fall within the red target area). **f**, Selection of the best TADF candidates using web voting. **g**, Synthesis of the selected molecules and testing as emitters in OLEDs. The OLED image shown in the inset of **g** is adapted from ref. 12, Nature Publishing Group.

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  $T_1$  ( $\Delta E_{ST}$ ) 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  $\Delta E_{ST}$  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  $\Delta E_{ST}$ ). 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  $\Delta E_{ST}$ , 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  $\Delta E_{ST}$  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  $\Delta E_{ST}$  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 applications<sup>11</sup>. 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.  $\square$

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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.

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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 flocks<sup>1</sup> or vortices<sup>2</sup>. 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 systems<sup>3</sup>. The second direction investigates novel active materials