Self-organized lasers from reconfigurable colloidal assemblies

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Non-equilibrium assemblies, where units are able to harness available energy to perform tasks, can often self-organize into dynamic materials that uniquely blend structure with functionality and responsiveness to their environment. The integration of similar features in photonic materials remains challenging but is desirable to manufacture active, adaptive and autonomous photonic devices. Here we show the self-organization of programmable random lasers from the reversible out-of-equilibrium self-assembly of colloids. The random lasing originates from the optical amplification of light undergoing multiple scattering within the dissipative colloidal assemblies and therefore depends crucially on their self-organization behaviour. Under external light stimuli, these dynamic random lasers are responsive and present a continuously tuneable laser threshold. They can therefore reconfigure and cooperate by emulating the ever-evolving spatiotemporal relationship between structure and functionality that is typical of many non-equilibrium assemblies.

Self-organization is the spontaneous emergence of structure and coordination from elementary units on larger scales than those defining the individual components. Many biological and artificial systems capable of harnessing available energy rely on this process to form complex structures and patterns as well as to achieve complex functionalities\(^1\). Inspired by their biological counterpart, the goal of controlling artificial non-equilibrium systems to self-assemble into reconfigurable, adaptive and autonomous artificial materials has driven a particularly vast scientific effort\(^2,3\). In colloidal science, colloidal systems that can engage in dissipative self-assembly after energy conversion have come to the fore due to their ability, for example, to emulate biological self-organization\(^4,5\), to transport and reversibly assemble passive colloidal cargoes\(^6–12\) and to implement microscopic metamachines and mechanical devices\(^13–20\).

Because of their easy synthesis with sizes comparable to the wavelengths of visible light, colloids have often been the building blocks of choice for photonic materials and devices with optical properties defined by their fixed topology and spatial correlations\(^21,22\). Adding optical gain to these static photonic assemblies can trigger lasing\(^23\). In disordered assemblies, random lasing\(^23–24\) has been observed in solid photonic glasses\(^25\), titania (TiO\(_2\)) colloidal systems\(^26\), semiconductor powders\(^27\) and more complex geometries\(^28,29\). These lasing functionalities emerging in the final assembled photonic materials are reaching technological applications (from low-coherence imaging\(^30\) to super-resolution spectroscopy\(^31\), from sensing\(^32\) to even interfacing with living tissues\(^33\)), thanks to the flexibility and shape insensitivity of random lasing processes.

Reconfigurable photonic architectures, capable of actively controlling light flow, are sought after for the implementation of next-generation photonic devices\(^34\), and reconfigurable lasers, whose lasing action can be controlled post-fabrication, are an emerging concept still in its infancy\(^35\). Random lasers fabricated from colloidal assemblies are indeed locked in their final configuration, thus proposing optical functionalities which are overwhelmingly static and fixed. Here, we realize programmable random lasers, which self-organize from the dissipative self-assembly of colloids after light absorption from a few units and show dynamic features such as responsiveness, reconfigurability and cooperation. Lasing emerges when the colloidal cluster dynamically reaches a threshold size which is controlled by the self-assembly process.

Colloids in solutions of laser dyes can scatter and amplify light that is trapped within them. When optically pumped by a high-energy laser of constant spot size and intensity (Methods), lasing can emerge when the local colloidal density increases above a threshold such that light travels an average optical path long enough for net amplification to occur before leaving the medium. This is the onset of random lasing\(^36\) reached by increased scattering. In Fig. 1a,b, we drive the self-assembly of freely diffusing polyethyleneimine-functionalized monodisperse TiO\(_2\) colloids with radius \(R_{\text{TiO}_2} = 0.915 \pm 0.03\, \mu\text{m}\) (Extended Data Figs. 1 and 2) in an ethanol solution of a rhodamine-based dye (rhodamine 6G or B) by generating a local temperature gradient around a carbon-coated Janus particle \((R_s = 4.22 \pm 0.14\, \mu\text{m})\) (Methods). By exploiting different mechanisms including, for example, convection, thermophoresis, thermo-osmosis, thermo-electricity and depletion effects\(^36–41\), light-induced temperature gradients are indeed well versed to manipulate colloidal particles and have found extensive use for optofluidic applications\(^32\). Under illumination by a continuous-wave 632.8 nm HeNe laser (Extended Data Fig. 3 and Methods), the Janus particle heats up because of light absorption, a TiO\(_2\) colloidal cluster assembles around it (Fig. 1b) and the particle assumes a cap-down orientation due to its equilibrium rotational dynamics (Extended Data Fig. 4). On removal of the external energy source, the colloids disperse reversibly (Fig. 1c). The process of dissipative gathering can then be reiterated, leading to re-accumulation of colloids around the Janus particle (Extended Data Fig. 5). As soon as a dense cluster is formed (Fig. 1b), lasing action can emerge dynamically via this phenomenon of dissipative self-organization. The lasing process is quantifiable by instantaneously measuring the emission spectra at different stages of this process (Fig. 1d) by pumping optically with a 532 nm pulsed laser (400 ps duration) at increasing pump fluence (Extended Data Fig. 3 and Methods). The lasing threshold is reached when the linewidth of the emission spectrum narrows to...
13.5 nm, that is, half of its initial value (Fig. 1e,f). Lasing emission is generated by optical modes extending in the plane containing the colloids, and we detect the light scattered in the out-of-plane direction. These spectra highlight the programmable optical functionality of these colloidal assemblies. Lasing can be switched on or off dynamically by controlling the density and size of the cluster within the pump region (52 μm in diameter). The initial concentration of $2 \times 10^{15}$ particles m$^{-3}$ (Fig. 1a) is too low to obtain lasing, and we only observe the broad emission characteristic of the dye fluorescence. A significantly larger, higher-density ($12 \times 10^{15}$ particles m$^{-3}$) cluster, as after accumulation and re-accumulation (Fig. 1b and Extended Data Fig. 5), however, shows a single narrow peak at 560 nm with linewidth of ~5 nm. During dispersal (Fig. 1c), the spectrum broadens again, thus evolving towards the initial fluorescence background. The power dependence evolution of the spectra in Fig. 1e,f indicates that the cluster is lasing at a threshold power of 70 mJ cm$^{-2}$, with two clear signatures of lasing: a marked superlinear increase in the emission intensity and a significant reduction in spectral linewidth.

We can interpret and quantitatively reproduce the dissipative accumulation of colloids, which leads to crossing the lasing threshold, by using a two-dimensional model based on thermal effects, where colloids in solution are drawn towards higher temperatures. Figure 2a shows the calculated steady-state temperature profile around the heat source (the illuminated Janus particle), which approximately decays with the inverse of the radial distance $r$ from the source (Methods). For the fixed HeNe laser intensity in our experiments (0.14 mW μm$^{-2}$), we measured a temperature...
Fig. 2 | Dynamics of dissipative colloidal accumulation and random lasing. a, b, The calculated temperature profile around a heat source (yellow circle) with the corresponding temperature-induced velocity field (arrows) for a TiO₂ colloid (cyan circle) (a). The overlaid simulated trajectory (magenta line) shows how the motion becomes more directed when approaching the heat source, consistent with the experimental (points) and modelled (line) colloidal radial velocity with distance \( r \) from the heat source (b). The error bars around each data point represent one s.d. around the mean values. c, The time dynamics of cluster formation (shaded area: experiments; dashed line: simulation) when the heat source is on (accumulation), off (dissipation) and on again (re-accumulation). The time points (circle and square) correspond to the start and end of accumulation. d, e, The cluster linewidth versus the particle density and pump radius for random laser theory (d) and experiments with rhodamine 6G (rhodamine B in Extended Data Fig. 8) (e). The dots highlight experimental values obtained at threshold (d) and at different linewidths (e) (values in nanometres in the plot) for different pump radii under a fixed pump fluence (140 mJ cm⁻²). The circle and square in d show the initial and final particle densities around the heat source before and after accumulation (as in c). The increase in particle density through accumulation explains the transition from below threshold (blue region) to lasing (red region).

The random lasing action occurs for a strongly enough scattering medium with a short enough scattering mean free path \( \ell_s \) and large enough excited cluster (as in Fig. 1b). This condition can be quantified by the critical radius \( R_\text{cr} \), a function of \( \ell_s \) as lasing action can only be achieved if the excited random laser area is of radius \( R_s > R_\text{cr} \) (ref. 27). For colloidal assemblies, \( R_s \) can be calculated by solving the radiative transfer equation for light (Methods). In our experiments, \( R_s \) is a time-dependent variable as \( \ell_s \) changes in time due to the accumulation and dispersal of colloids. Figure 2d reveals how \( R_s \) decreases monotonically with increasing particle densities, thus facilitating the narrowing of the linewidth and lasing action. This trend is confirmed by the equivalent experimental data in Fig. 2c, where we determine the dependence of \( R_s \) on the particle density by pumping large colloidal assemblies with different TiO₂ concentrations at a fixed pump fluence (140 mJ cm⁻²) while varying the pump spot size (Methods). On the basis of Fig. 1c,f, \( R_s \) is taken as the pump size at which the linewidth of the emission spectrum narrows to half of its initial value, that is, to 13.5 nm, in good agreement with model predictions (Fig. 2d), where the only fitting parameter is the net gain length (Methods). The particle densities before and after accumulation are marked on Fig. 2d, showing that \( R_s \) is smaller (larger) than \( R_\text{cr} \) before (after) accumulation, thus decreasing of \( \Delta T = 57 ± 1.6^\circ \text{C} \) over room temperature, corresponding to a source temperature of \( T_s = 78 ± 1.6^\circ \text{C} \) (Extended Data Fig. 6). This sharp radial thermal gradient produces a temperature-induced drift, which drags the TiO₂ particles in the radial direction determined by their overall temperature-induced mobility \( \mu_r \). This parameter incorporates all the information about the interfacial interactions between the liquid and the solid, which may be quite complex owing to, for example, thermo-osmotic, thermophoretic, thermo-electric or depletion contributions 41,42. The drift velocity of the colloids in the radial direction \( \mathbf{v}_r \), can then be defined as 41,42.

\[
\mathbf{v}(r) = -\mu_r \frac{\partial T(r)}{\partial r} \mathbf{e}_r
\]

where \( \mu_r \approx 1 \text{ m}^2 \text{K}^{-1} \text{s}^{-1} \) is the only fitting parameter, having the units of a thermo-diffusion coefficient, extracted from our data (Fig. 2b). As \( \mu_r < 0 \), particles migrate towards warmer regions. Figure 2b and the arrows in Fig. 2a show how the magnitude of \( \mathbf{v}(r) \) increases, in both experiments and the model, as \( r \to 0 \) when nearing the heat source, so that, as shown by the simulated trajectory in Fig. 2a (Methods), the motion of a single TiO₂ particle becomes more directed with proximity to the source. The experimental dissipative dynamics of accumulation, dispersal and re-accumulation of the colloidal assembly can thus be well replicated with a simple two-dimensional particle-based model that includes this temperature-induced drift towards the heat source and a short-range repulsive interaction among the colloids on the order of \(-k_BT\) (Fig. 2c and Methods). The rate of growth of the cluster is steeper at the start of the process as colloids within a distance \( \sim 4R_s \) from the Janus particle experience stronger drifts (>1 μm s⁻¹; Fig. 2b). As nearby colloids are drawn quickly towards the heat source, those that are farther away take longer to arrive as their motion is dominated by their Brownian dynamics (less directed), because \( \mathbf{v} \to 0 \) as \( \partial T/\partial r \to 0 \) (that is, for \( r \to \infty \)), thus the rate of accumulation of new colloids around the cluster slows down in time 41. When the heat source is turned off, the assembly dissolves, driven by diffusion and a short-ranged interparticle repulsion with a rate of dispersal tending to zero as the colloids evolve towards an equilibrium distribution (Fig. 2c and Methods). When the heat source is back on, colloids re-accumulate much faster than in the first accumulation phase due to the now higher colloidal density around the heat source (Fig. 2c and Extended Data Fig. 5).
explaining why we observe lasing only after a certain accumulation time, that is, for large enough assemblies (larger than $R_c$), yet not before accumulation or after dissipation.

The dynamic nature of the dissipative self-organization behind our random lasers, linking structure with optical functionality, can be harnessed to achieve unconventional tasks for standard random lasers such as reconfigurability in space and time. Employing heat sources at different locations, lasing action can be triggered first and then transferred in space, thus shifting the lasing load within the sample. Figure 3 shows how two Janus particles, placed approximately one pump spot size apart, alternately act as accumulation points for the TiO$_2$ colloids when respectively activated by the external energy source. The lasing action is then switched from one heat source to the other by transferring the colloidal scattering load across and back.

Janus particles can also cooperate to achieve feats beyond what is achievable using a single particle. Figure 4 shows how the cooperation among properly located heat sources can lead to a boost in...
the lasing properties and morph the laser spatially by imparting different shapes to the colloidal assemblies. While the colloidal assembly in Fig. 4a is too small to lase as \( R_c < R_{cr} \) (Fig. 4b), the addition of further colloids by a second Janus particle (Fig. 4c) pushes the cluster size just above the threshold for lasing \( (R_c \approx R_{cr}, \text{Fig. 4d}) \). This effect becomes even more pronounced when a third Janus particle joins the assembly, as \( R_c > R_{cr} \) (Fig. 4c,d). Beyond the possibility of refining the laser spectral properties, another alluring application for multiple cooperating Janus particles is the possibility of defining different planar laser geometries (Fig. 4e), which could be important for display applications, for travelling through narrow channels or for adhering to complexly shaped targets.

In conclusion, we have performed the first experimental demonstration of an artificial colloidal material that, by virtue of dissipative self-assembly dynamics, can spontaneously self-organize in a random laser device, which dynamically blends morphology with its optical functionality. By reversibly accumulating colloids with temperature gradients, we enable an advanced photonic application (that is, lasing), when, previously, particle motion in thermal gradients had only been used to inhibit lasing\(^1\). While here we employed Janus particles as heat sources to control the colloidal self-assembly dynamics, our results can be generalized to alternative light-absorbing materials (for example, other gold- and carbon-based materials or dye-doped beads) as well as to direct light absorption by the solvent, provided that these alternative approaches do not interfere with the laser action (for example, by causing dye bleaching effects). These self-organized random lasers can be manipulated on demand to produce controllable and programmable lasing, thus bringing novel functionalities to the field of photonics and opening the door to a new class of active functional materials in active matter. Due to the efficient gain medium and strong light trapping properties, these lasers have a relatively small size (a few tens of microns), which can be beneficial for applications requiring small footprints, including the realization of display pixels, high-brightness light sources for low-coherence illumination, sensors for microfluidics and signal multiplexers\(^5\). While the lasers demonstrated here are quasi-two-dimensional and require minutes to reconfigure, two- and three-dimensional random lasers with faster responses (albeit still slower than electronic switching) can be achieved by, for example, exploiting additional or alternative stimuli to thermal gradients, such as optical or electric fields\(^5,46\), to find application in, for example, e-ink displays. Ultimately, the responsiveness, reconfigurability and cooperation properties of our self-organized lasers are a first step towards the realization of fully animate lasers capable of independent motion and autonomous adaptation in response to external stimuli. Indeed, we envisage that the realization of similar self-organized lasers from light-actuated colloidal molecules\(^46,47\) will pave the way towards the development of a new class of functional materials with potential for sensing applications\(^47\), non-conventional computing\(^48\), novel light sources, speckle-free illumination\(^49\) and display technology\(^49\).

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at [https://doi.org/10.1038/s41567-022-01656-2](https://doi.org/10.1038/s41567-022-01656-2).

Received: 18 January 2022; Accepted: 27 May 2022; Published online: 14 July 2022

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Methods

Materials. Glass microscopy slides (Thermo Fisher) were purchased from VWR, while glass coverslips were purchased from Thorslab. The following chemicals were purchased and used as received: rhodamine 6G (Sigma-Aldrich), rhodamine B (Acros Organics), acetone (≥99.8%, Sigma-Aldrich), ethanol (≥99.8% Fisher Scientific), sodium hydroxide (NaOH; Fisher Scientific) and polyethyleneimine (PEI, branched, Mw 25,000, Mn 10,000; Sigma-Aldrich). Deionized (DI) water (≥18 MΩ cm) was collected from a Milli-Q purification system. Aqueous colloidal dispersions (5% w/v) of silica (SiO2) colloids for fabricating Janus particles and to be used as spacers (8.44 ± 0.27 μm and 20 ± 0.64 μm in diameter, respectively) were purchased from Microparticules GmbH. Aqueous colloidal dispersions of titania (TiO2) particles (2.5% w/v, 1.73 ± 0.03 μm in diameter) for the lasing experiments, of fluorescent SiO2 colloids (2.5% w/v, 10.05 ± 0.31 μm, excitation/emission 602 nm/623 nm) for Extended Data Fig. 4 and of polystyrene particles (10% w/v, 1.65 ± 0.04 μm) for Extended Data Fig. 7 were also purchased from Microparticules GmbH. Carbon rods with length of 300 mm and diameter of 6.15 mm for coating Janus particles were purchased from Agar Scientific and cut to a length of 50 mm before use. Ultraviolet-cure adhesive (Blufixx) and hydrophobic coating (RainX) for sample preparation were purchased from an online retailer (Amazon).

Slide cleaning protocol. Before their use for sample preparation, glass slides and coverslips were cleaned via sonication for 10 min in 2 M NaOH ethanolic solution, followed by three cycles of 5 min of sonication in DI water. To dry them, the slides were withdrawn from the water in the presence of ethanol vapour (Marangoni drying) and, subsequently, blown with a nitrogen gun.

Fabrication of Janus particles. The Janus particles used as heat sources in our experiments were fabricated from SiO2 colloids with radius $R_s = 4.22 ± 0.14 \mu m$, which were coated on one side with a thin layer (~60 nm) of carbon. We first deposited a monolayer of colloids on a clean glass slide. The monolayer was extended by evaporating a 40 μL droplet containing a 2.5% w/v dispersion of the colloids in DI water. The particles were then coated with a 60 nm-thick carbon layer using an automatic carbon coater (AGB7367A, Agar Scientific). Post-coating sonication allowed us to dislodge the half-coated particles in DI water from the glass slides to use them for sample preparation. Due to their micrometric size, the use of Janus particles as heat sources eased their visualization, characterization and manipulation in crowded colloidal environments. The ability to easily identify the heat sources was key for tracking them in Figs. 3 and 4 and for measuring the induced local temperature increase as a function of laser power in Extended Data Fig. 6.

Preparation of samples of colloids and laser dyes. Random lasers combine optical gain with a scattering medium. The samples used are in the form of a glass chamber (see next section) filled with a colloidal dispersion in an ethanol solution of laser dye. The optical gain is provided by two types of rhodamine-based dyes in ethanol solution (1% w/v): rhodamine 6G (R6G) for Figs. 1 and 2 and Extended Data Figs. 5 and 7, and rhodamine B (Rhb) for Figs. 3 and 4 and Extended Data Fig. 8. The use of the two dyes is motivated by their complementary features in experiments. At $\lambda = 532 \text{ nm}$ (the wavelength of the pump laser), pumping rhodamine 6G (absorption peak at 530 nm) is more efficient than pumping rhodamine B (absorption peak at $\lambda = 550 \text{ nm}$), thus improving the lasing performance at a given power (that is, narrower linewidth). Rhb solutions instead reduce the sticking of Janus particles to the glass substrate, thus performing better for tasks where their manoeuvrability is paramount. The scattering medium is a dispersion of monodisperse TiO2 colloids, chosen due to the material’s characteristic high refractive index (nTiO2 ≈ 2.3 for amorphous titania), higher than that of ethanol (nethanol = 1.36). This refractive index difference is important to achieve the strong scattering properties needed for lasing action. In fact, no lasing is observed when substituting the TiO2 colloids with lower-refractive-index polymer colloids of a similar size (Extended Data Fig. 7). We obtained stable dispersions of TiO2 colloids in 1% w/v ethanol solutions of laser dyes (either rhodamine 6G or rhodamine B) by functionalizing the colloids with PEI to prevent flocculation and sticking to the glass substrate. In particular, we first mixed 50 μL of 0.1% aqueous dispersion of Janus particles with stock solutions of TiO2 colloids (20 μL) and 20 μm SiO2 colloids (10 μL) in a 1.5 mL centrifuge tube (Eppendorf).

Sample chamber preparation. Experiment-ready sample chambers containing a dispersion of colloids in ethanol solutions of laser dyes were prepared by sandwiching 15 μL of the dispersion between a clean glass slide and a thin coverslip using concentrations of 20 μm TiO2 colloids as spacers. The slide and coverslip were soaked for 2 min in Rain X, a commercial solution which renders glass surfaces more hydrophobic and aids limiting particle sticking to the glass chamber. Excess RainX was removed by soaking the slide in acetone and subsequently wiping with lens tissue. The chamber was then sealed by applying an ultraviolet-cure adhesive around the borders of the coverslip, taking care not to expose the dye solution to ultraviolet light by illuminating only the edges of the coverslip, as this could cause dye bleaching. Before data acquisition, the sample was left to equilibrate over a 1 h period.

Optical setup and microscopy. Extended Data Fig. 3 shows a schematic of the experimental setup used to illuminate the Janus particles, to image the sample and to probe its emission spectra. Samples are mounted on the stage of a Nikon Ti microscope. Two laser sources are exploited: a continuous-wave HeNe laser (Thorlabs, λ = 632.8 nm, 20 mW) and a Nd:YAG pulsed laser (TEEM Power-Chip, λ = 532 nm, pulse width 400 ps, energy per pulse 20 μJ, 1–10 kHz). The HeNe laser is used as an energy source for heating the Janus particles, while the pulsed laser is used to reach population inversion for lasing measurements. The choice of using the HeNe as energy source was dictated by the need to avoid overlap with the absorption spectrum of rhodamine dyes (centred at 525 nm), thus avoiding spurious heating effects in the sample due to this competing absorption process in the dye. The pulsed laser is only operating when measuring emission spectra and in the lasing mode for less than 90 s at each time to limit absorption from the Janus particle and to avoid modification of the accumulated colloidal cluster.

The HeNe laser is coupled to a single-mode optical fibre, and the beam from the fibre is focused onto the sample with a lens of 60 mm focal length to a spot diameter of ~5 μm. This spot size was chosen to match the size of the Janus particle while keeping the laser on it while making sure that the HeNe laser was only operating with sufficient power density. For accumulation of TiO2 colloids around the Janus particle to occur, the laser power density at the sample was around 0.14 mW μm−2. The laser power was controlled by a movable knife edge before the fibre couple.

The same HeNe laser was also used to manipulate the Janus particles by optical forces or cavitation (see next section).

For lasing measurements, samples were optically pumped at room temperature with the Nd:YAG pump laser. The pump laser profile was shaped with a programmable digital micro-mirror device (DMD, Ajile AJD–4500), and the excitation pattern was imaged onto the sample through a 40x objectiﬁc lens (Nikon CF Plan Fluor 40x, 0.75 N.A., 0.66 mm W.D.). An acousto-optic modulator is used to control the energy of the pump laser. Circular illumination proﬁles, of constant intensity in a disk shape and radius $R_{\text{circular}}$, were used for the measurements in Figs. 1, 2 and 4 and Extended Data Figs. 5 and 8. Doughnut-shaped proﬁles, of constant intensity in a disk shape without the central part, centred on the Janus particle, were used for Fig. 3 and Extended Data Fig. 7 to avoid further laser exposure and prevent any motion of the Janus particle. The size of the missing central part was chosen to be about 1.5 times larger than the size of the Janus particle, for ease of alignment. When the sample was pumped, the HeNe laser was blocked to avoid overheating the Janus particle, and in particular to minimize the formation of cavitation bubbles during the lasing measurements. The laser energy detected from the sample was absorbed through the same objective lens, filtered and then focused into a stripe on the spectrometer entrance slit via a cylindrical lens to maximize the measured signal counts. The signal was spectrally analysed using a grating spectrometer (Princeton Instruments Isoplane-320) equipped with a 600 gr mm−1 visible grating (0.5 nm resolution) and a charge-coupled device camera (Princeton Instruments Pixis 400). Linewidths are given as full width at half maximum. Fluctuations in the recorded spectra are caused by the dynamic colloidal system as well as by the small fluctuations (around 3%) of the pump laser pulse energy. Overall, the variation of the intensity peak of the random lasing spectra is of approximately 6.4%, which is the s.d. obtained from a 6-min-long measurement.

The critical radius $R_c$, in Fig. 2 was measured at a constant pump ﬂuence of 140 mW cm−2, by changing the size of the illumination spot of radius $R_{\text{in}},$ while recording the emission spectrum, until the emission linewidth reached the value of 13.5 nm, which is half the fluorescence linewidth measured at low pumping powers.

The motion of the TiO2 colloids, in Figs. 1 and 2 and Extended Data Fig. 5 was recorded by using a complementary metal–oxide–semiconductor camera (Thorlabs) at a frame rate of 2 frames per second. The image focus was adjusted so that each particle had a bright spot at its centre relative to the background, to provide enough contrast to discern individual particles via digital video microscopy based on homemade MATLAB tracking software.

Manipulation of Janus particles. To position the Janus particles at different locations in the sample chamber, we manipulated them with the mildly focused HeNe laser, which, at power densities of 0.14 mW μm−2, exerts a gentle pulling optical force which predominantly drags the Janus particle towards the centre of the laser spot. The manipulation was performed by either moving the sample stage...
only (Figs. 1, 2 and 3 and Extended Data Figs. 5 and 7) or the HeNe laser spot only (Fig. 4), while keeping the other element fixed. The same HeNe laser at higher power densities (above 0.2 mW μm⁻²) was also used to free the Janus particles in TiO₂ clusters by cavitating before repositioning them (Figs. 1 and 3). Cavitating is created when the Janus particle is strongly heated by the HeNe laser, leading to bubble formation (Extended Data Fig. 6) and provides an instantaneous pushing action against the carbon side of the Janus particle that propels it away from the surrounding TiO₂ colloids.

Calculation of the temperature profile around a heat source. As the dynamics of our dissipative colloidal assemblies around an illuminated Janus particle predominantly take place near the glass surface, we can calculate the temperature profile generated by a disc heat source of radius $R$, at temperature $T$, in two dimensions. This is a reasonable approximation considering that the most likely configuration for the Janus particle in a formed cluster is with the cap facing down, that is, towards the interface (Extended Data Fig. 4). Under continuous illumination, the heating of the surrounding fluid can be assumed to be instantaneous so that a steady-state temperature profile around the heat source is promptly reached. In fact, since heat propagation is much faster than particle migration, the temperature field can be considered as stationary. Assuming a steady state for the diffusion of heat from the source, we can then calculate the temperature profile around the heat source in the plane of motion as

$$T(r) = \frac{2}{\pi} \sum_i \left( T_i - T_0 \sin^{-1} \left( \frac{R_i}{r} \right) + T_0 \right), \quad r \geq R_i,$$

where $r$ is the radial distance from the centre of the heat source and $T_i$ is room temperature. Note that, in steady state, the presence of an interface does not influence the temperature profile.

Particle-based simulations. We consider a simple numerical model where $N$ hard spheres of mean radius $R_{TiO2}$ move inside a two-dimensional square box of side $\beta = 187 \mu m$. Particles were placed at random without overlap at a fixed density ($0.0425$ particles $\mu m^{-3}$). Unless otherwise specified, the values for all the parameters in the simulations are set to the exact experimental values reported in the main text. The radius of each individual particle $R_i$ is taken from a Gaussian distribution with mean $R_{TiO2}$ and standard deviation $\delta R$ to reproduce the size variability of the monodisperse sample of TiO₂ colloids. At the centre of the box, we place a disc heat source of radius $R$. When the source is active, it has a uniform temperature $T_s$ and generates a radial temperature profile according to equation (2). When the source is not active, the temperature is uniform in the box and equivalent to room temperature $T_0$.

The trajectory of the $i$th particle is then obtained by solving the following Langevin equation in the overdamped regime using the second-order stochastic Runge–Kutta numerical scheme:

$$\dot{x}_i = u_i + \sum_{j \neq i} \frac{F_{ij}}{T_j} + \sqrt{2D} \xi_i,$$

where $x_i$ and $u_i$ are respectively the particle's position and temperature-induced drift velocity at time $t$ (equation (1)), $D = k_B T_s / \gamma$ is the particle's diffusion coefficient at position $x_i$ and temperature $T_s$ (equation (2)) with $\gamma$ its friction coefficient and $\xi_i$ is a two-dimensional vector of an independent white noise process with zero mean and unitary variance. When the heat source is off, $u_i$ is null. The direction of motion due to the temperature-induced drift velocity is therefore defined by the unitary vector $\vec{\xi}(t) = [\cos(\theta(t)), \sin(\theta(t))], \theta(t)$ is the particle's angular coordinate in the frame of reference defined by the heat source.

We implemented particle–particle sterics $F_{ij}$ with the repulsive term of a Lennard–Jones potential with parameters $e = k_B T_0$ and $\sigma = 2R_{TiO2}$. The effect of this repulsive term is short ranged and was truncated at $1.5(R_i + R_j)$ for each pair of neighbouring particles $i$ and $j$. Finally, we modelled the steric interaction with the heat source by introducing a repulsive force $F_e(r)$ into the equation of motion. This force depends on the particle's distance $r$, from the heat source as

$$F_e(r) \propto \frac{e^{-\gamma}}{|r_t - R_s + R_i|}.$$

This function was chosen to reproduce a strong (local) repulsive interaction between particle and source, that is, to mimic a hardcore potential. The exponential term ensures that the force does not increase too abruptly when approaching the source. This effect of this force was truncated at a cut-off distance of $r_t = 2.5 R_s$.

Random lasing model. The random lasing model is based on the radiative transfer equation - and estimates the critical radius $R_c$, that is, the minimum size of a colloidal cluster needed to achieve the lasing threshold. Since the lateral dimensions of our sample are much larger than its thickness, we used a two-dimensional model where $R_c$ is given by solving

$$\frac{j_0 \left( \frac{2 g (\ell^2 - g) R_c}{} \right)}{j_1 \left( \frac{2 g (\ell^2 - g) R_c}{} \right)} = \frac{\pi}{2 g (\ell^2 - g)}.$$

where $j_0$ and $j_1$ are Bessel functions of the first kind, $\ell_c = 1/(\sigma r_0)$ is the scattering length (where $\sigma_s$ is the scattering cross-section and $r_0$ is the particle density) and $g = 1/\ell_c^2$ is the gain coefficient of the gain medium, with $\ell_c$ being the dye gain length. In our self-organized lasers, $\ell_c$ and hence $R_c$, depend on the time-varying particle density in the accumulation/dispersal processes.

To determine the critical radii of the clusters with different particle densities at different times, equation (5) was solved numerically, with $g$ a fitting parameter and $\ell_c$ obtained from Mie theory for TiO₂ colloids of 1.83 μm diameter and a refractive index of $n_{TiO2} = 2.3$. We chose to fit $g$ because its exact value depends on off-plane scattering losses (that is, when light escapes from the sample plane) and on minor absorption losses from the partially pumped dye and the Janus particle, thus usually being larger than $\ell_c$. A good fit to the experimental data in Fig. 2d is obtained with $g_{\text{cont}} = 0.0721$ μm⁻¹ (corresponding to a net dye gain length of $\ell_c = 13.87$ μm). This gain length estimate is around 3.5 times larger than $\ell_{\text{cont}} = 4$ μm given that, for rhodamine, $\sigma_{\text{cont}} = 2 \times 10^{-21}$ m² (ref. 24) and that the dye density is $\rho_d = 1.257 \times 10^{-24} \text{molecules m}^{-3}$ in a 1% w/v ethanol solution (the molar mass of rhodamine being 679 g mol⁻¹).

The value of $\ell_c$ (as estimated from Mie scattering calculations and particle counting from bright-field images) reaches a minimum value of 13 μm for a dense cluster, much longer than the light wavelength, while for a dilute cluster it is on the order of 100 μm. The optical thickness, that is, the cluster diameter divided by $\ell_c$, is smaller than 6. This justifies our modelling of the random laser in the intermediate regime between the diffusive and ballistic limits.

Data availability
Source data are available for this paper in Figshare with the digital object identifier 10.6084/m9.figshare.19745293 (https://doi.org/10.6084/m9.figshare.19745293). All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Code availability
The code that supports the findings of this study is available from the corresponding authors upon reasonable request.

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Acknowledgements
We are grateful to S. Rueber, M. Blunt and V. Barbieri for initial training on experimental techniques. G.V. acknowledges sponsorship for this work by the US Office of Naval Research Global (award no. N62909-18-1-1270). W.K.N. acknowledges the research support funded by the President’s PhD Scholarships from Imperial College London. R.S. and D.S. acknowledge support from The Engineering and Physical Sciences Research Council (EPSRC), grant no. EP/I027258, and the European Community.

Author contributions
Author contributions are defined based on the CRediT (Contributor Roles Taxonomy) and listed alphabetically: Conceptualization: R.S. and G.V. Data curation: W.K.N. and M.T. Formal analysis: W.K.N., D.S., M.T. and G.V. Funding acquisition: R.S. and G.V. Investigation: W.K.N., R.S., D.S., M.T. and G.V. Methodology: W.K.N., R.S., M.T. and G.V. Project administration: R.S., D.S., M.T. and G.V. Software: W.K.N., D.S. and G.V.
Supervision: R.S. and G.V. Validation: W.K.N., D.S. and M.T. Visualization: All. Writing–original draft: All. Writing–review and editing: All.

Competing interests
The authors declare no competing interests.

Additional information
Extended data is available for this paper at https://doi.org/10.1038/s41567-022-01656-2.

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41567-022-01656-2.

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Peer review information Nature Physics thanks Neda Ghotfaniha and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Diffusion of TiO$_2$ colloids in ethanol solutions of rhodamine dyes. In our experiments, due to their size and non-negligible density ($\rho_{\text{TiO}_2} = 4.24$ g cm$^{-3}$), TiO$_2$ colloids quickly sediment at the bottom surface of the experimental chamber, where, in the absence of a heat source, they primarily diffuse in 1% w/v ethanol solutions of rhodamine dyes near the plane defined by this interface. The diffusive behaviour is confirmed by the ensemble-averaged mean squared displacements (MSD(\tau) \propto \tau) for both rhodamine 6G (circles) and rhodamine B (triangles). Each MSD is calculated from the trajectories of at least 6 colloids. The experimentally measured diffusion coefficients are $D = 0.071 \pm 0.002 \mu m^2 s^{-1}$ for colloids in rhodamine 6G and $D = 0.069 \pm 0.001 \mu m^2 s^{-1}$ for colloids in rhodamine B.
Extended Data Fig. 2 | Functionalisation of TiO<sub>2</sub> colloids. Fourier-transform infrared spectroscopy (FTIR) measurements of polyethylenimine polymer (PEI), pristine titania colloids (TiO<sub>2</sub>), and PEI-functionalised titania colloids (PEI@TiO<sub>2</sub>). Notable peaks at 1740 cm<sup>-1</sup>, 1365 cm<sup>-1</sup> and 1217 cm<sup>-1</sup> (dashed lines) corresponding to the presence of PEI are found on the functionalised TiO<sub>2</sub> colloids, but not on the pristine particles. Spectra (except that of PEI) are enhanced by a factor of 10 for an improved comparison.
Extended Data Fig. 3 | Experimental setup. Two lasers (Nd:YAG and HeNe) are incorporated into a microscope where the sample with the colloidal particles is held (inset). The HeNe laser is used as an illumination source for the Janus particles, while the pulsed laser is used for lasing measurements. The pump power is controlled by an acoustic optical modulator (AOM), and a digital micro-mirror device (DMD) in the optical path is used to shape its excitation spot. The fibre-coupled HeNe laser is mounted on a separate stage and placed at the top of the sample. The lasing emission from the sample is collected and filtered by a 532 nm dichroic mirror (DM) and two filters (532 nm long-pass and 600 nm short-pass), then spectrally analysed by a spectrometer. The lamp shown in the inset illuminates the sample for bright-field imaging. The sample image is then sent to a camera through a 30:70 beam splitter (BS). M: mirror; CL: cylindrical lens.
Extended Data Fig. 4 | Orientation of Janus particles within colloidal assemblies. Janus particles fabricated from fluorescent SiO$_2$ colloids (~10 μm in diameter, excitation peak at 602 nm; emission peak at 623 nm) were used to confirm the orientation of the Janus particles in our experiments after accumulation of TiO$_2$ colloids. (a-c) Schematics (top) and representative bright-field (middle) and fluorescence (bottom) images of the orientation of Janus particles under different conditions. (a) A Janus particle is initially found in a cap-up configuration near the bottom surface of the sample chamber. When excited at 620 nm from the same side of the image detection, it appears bright under fluorescence imaging. (b) After illuminating the particle in a with a continuous-wave laser at 532 nm to induce significant accumulation of TiO$_2$ colloids, the particle is found to assume a cap-down configuration. In fact, when its translational motion is hampered by the surrounding cluster, the particle’s diffusive rotational dynamics lead to a cap-down equilibrium orientation dictated by gravity, due to the higher density of carbon ($\rho_c = 3.52$ g cm$^{-3}$) with respect to silica ($\rho_{SiO_2} = 2.65$ g cm$^{-3}$). After having switched off the external illumination by laser light, the detected fluorescence for this particle is indeed visibly lower than in a. The difference in brightness is due to the light screening effect introduced when the carbon cap is facing downwards. (c) This is confirmed by another Janus particle which has sedimented at the bottom surface of the sample chamber and is observed to diffuse with the cap-down equilibrium orientation dictated by gravity. Similar to b, fluorescence light is screened by the cap, hence the particle appears darker under fluorescence microscopy. Imaging and illumination settings were kept constant for all measurements.
Extended Data Fig. 5 | Re-accumulation of TiO$_2$ colloids after dispersal. After dispersal (Fig. 1c), TiO$_2$ colloids can be re-accumulated around the Janus particle and lasing action restored (Fig. 1d). A much shorter time (~10 minutes) is needed to re-accumulate particles to the same level as at the end of the accumulation phase due to the increased colloidal density after the first round of accumulation and dispersal. For the same reason, lasing action can also be reinstated much faster (~10 times): lasing is first observed after ~100 s of re-accumulation versus ~1200 s during accumulation. A few 275 second-long trajectories highlight the colloids’ motion.
Extended Data Fig. 6 | Temperature increase measurement around a Janus particle. All our experiments were performed by illuminating a Janus particle at a fixed HeNe laser power density (0.14 mW μm⁻²) corresponding to a local temperature increase ΔT = 57 ± 1.6°C (cross), just below the boiling point of ethanol (T_b = 78.37°C). Above this power density, a cavitation bubble forms around the cap of the Janus particle (inset). To confirm this temperature increase, we calibrated the relationship between laser power density and ΔT by detecting the onset of the demixing of a critical mixture (2,6-lutidine and water) with respect to a set temperature as a function of laser power (circles). The sample containing the Janus particles for the measurements was placed on a homemade temperature stage with a precision of 0.1 K and allowed to equilibrate for 10 min at a range of different temperatures, before illuminating the particle with gradually increasing laser powers. Each of these temperatures correspond to a well-defined ΔT with respect to the critical temperature of the mixture (T_c = 307.25 K), thus allowing to univocally identify the corresponding power density at which demixing of the critical mixture is clearly observed. Fitting a linear trend to the whole set of data (dashed line) allows us to verify the reliability of our initial temperature estimation due to boiling in ethanol (cross). The error bars around each data point represent one standard deviation around the mean values.
Extended Data Fig. 7 | Absence of lasing in assemblies of low refractive index colloids. (a–b) A light-absorbing Janus particle in a laser dye solution with polymer (polystyrene, PS) microparticles attracts the diffusing colloids (accumulation) when illuminated by a HeNe laser (CW, 632.8 nm). (b) The colloids assemble in a dense cluster as in Fig. 1 for TiO$_2$ particles. The polystyrene colloids have similar size and initial concentration as the TiO$_2$ particles in Fig. 1. The dashed area in a–b represents the pump region (52 μm in diameter). (c) As scattering from the polystyrene particles is weaker than that from TiO$_2$ particles due to their lower refractive index, the spectra for the polymer colloids before and after accumulation show no lasing action. Nonetheless, a similar size cluster of TiO$_2$ particles is lasing (black dashed line). All spectra are obtained at the pump fluence of 130 mJ cm$^{-2}$. (d–e) The absence of lasing action from the polystyrene cluster is confirmed by the linear (rather than superlinear) increase of the peak intensity as a function of pump fluence in d and by the fact that the emission linewidth does not reduce below 13.5 nm (lasing threshold) in e.
Extended Data Fig. 8 | Characterisation of critical radius in TiO₂ colloidal samples with rhodamine B dye. We have performed experiments with both rhodamine 6G and rhodamine B (Methods). The measurement of the critical radius and its modelling has been performed for both dyes. Fig. 2d-e is done with rhodamine 6G, while this figure is done with rhodamine B. In both cases the colloidal samples are similar, and fabricated with the same procedure. The dots in a and b highlight experimental values obtained (a) at threshold and (b) at different linewidths (values in nm in the plot) for different pump radii under a fixed pump fluence (140 mJ cm⁻²). The lasing threshold (white) is defined when the full width at half maximum is 13.5 nm. The RTE model in a uses a gain length of 11.09 μm as the only free parameter to fit the experimental data (Methods). Theory and experiments show good agreement.