Supplementary Figures
Clustering of Janus Particles in Optical Potential Driven by Hydrodynamic Fluxes

S. Masoumeh Mousavi,[1] Iryna Kasianiuk,[2] Denis Kasyanyuk,[2] Sabareesh K. P. Velu,[1] Agnese Callegari,[2] Luca Biancofiore,[3] and Giovanni Volpe[1,4]

[1] *Soft Matter Lab, Department of Physics, Bilkent University, Ankara 06800, Turkey*
[2] *Soft Matter Lab, Department of Physics, Bilkent University, and UNAM, Ankara 06800, Turkey*
[3] *Department of Mechanical Engineering, Bilkent University, Ankara 06800, Turkey*
[4] *Department of Physics, University of Gothenburg, SE-41296 Gothenburg, Sweden*

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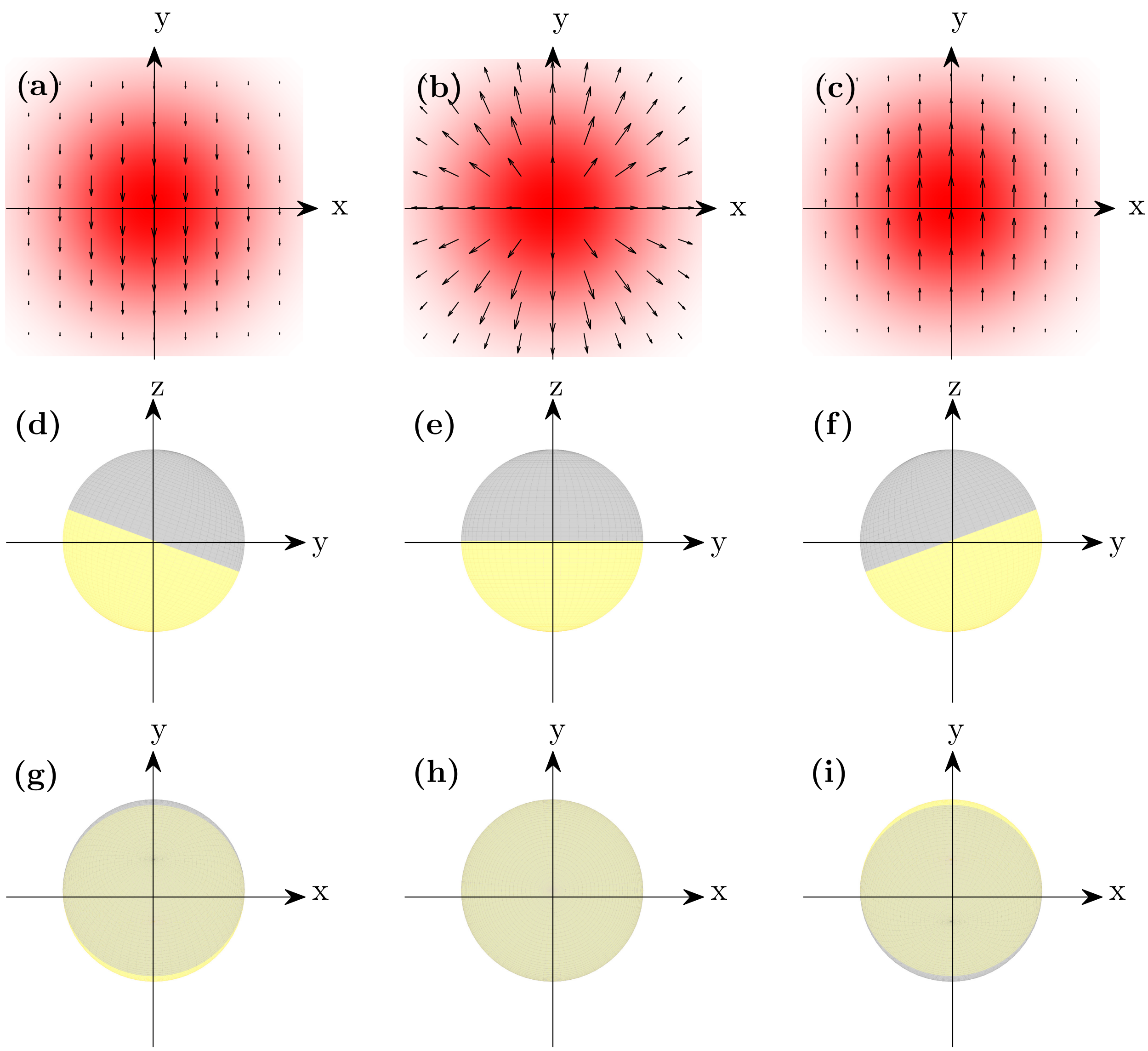
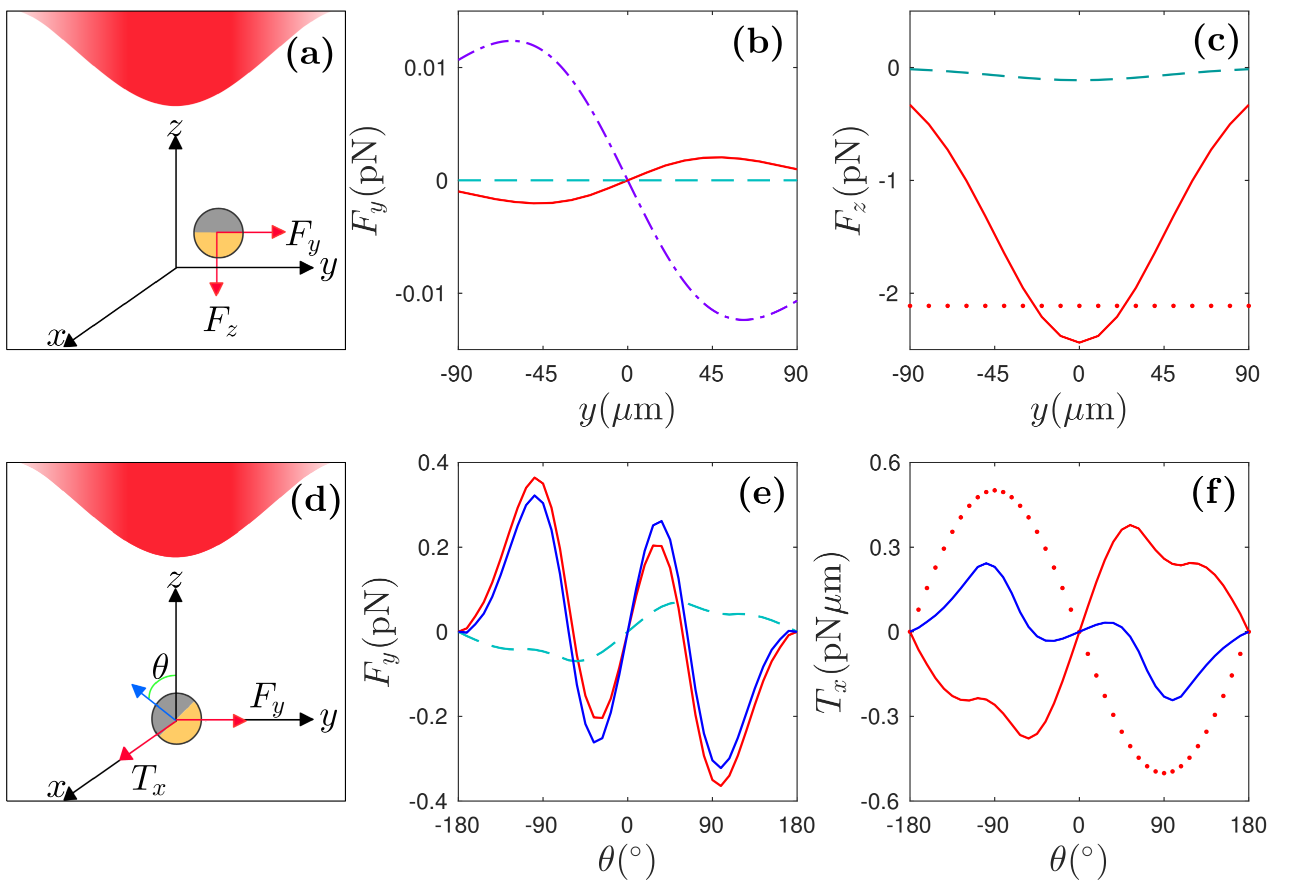
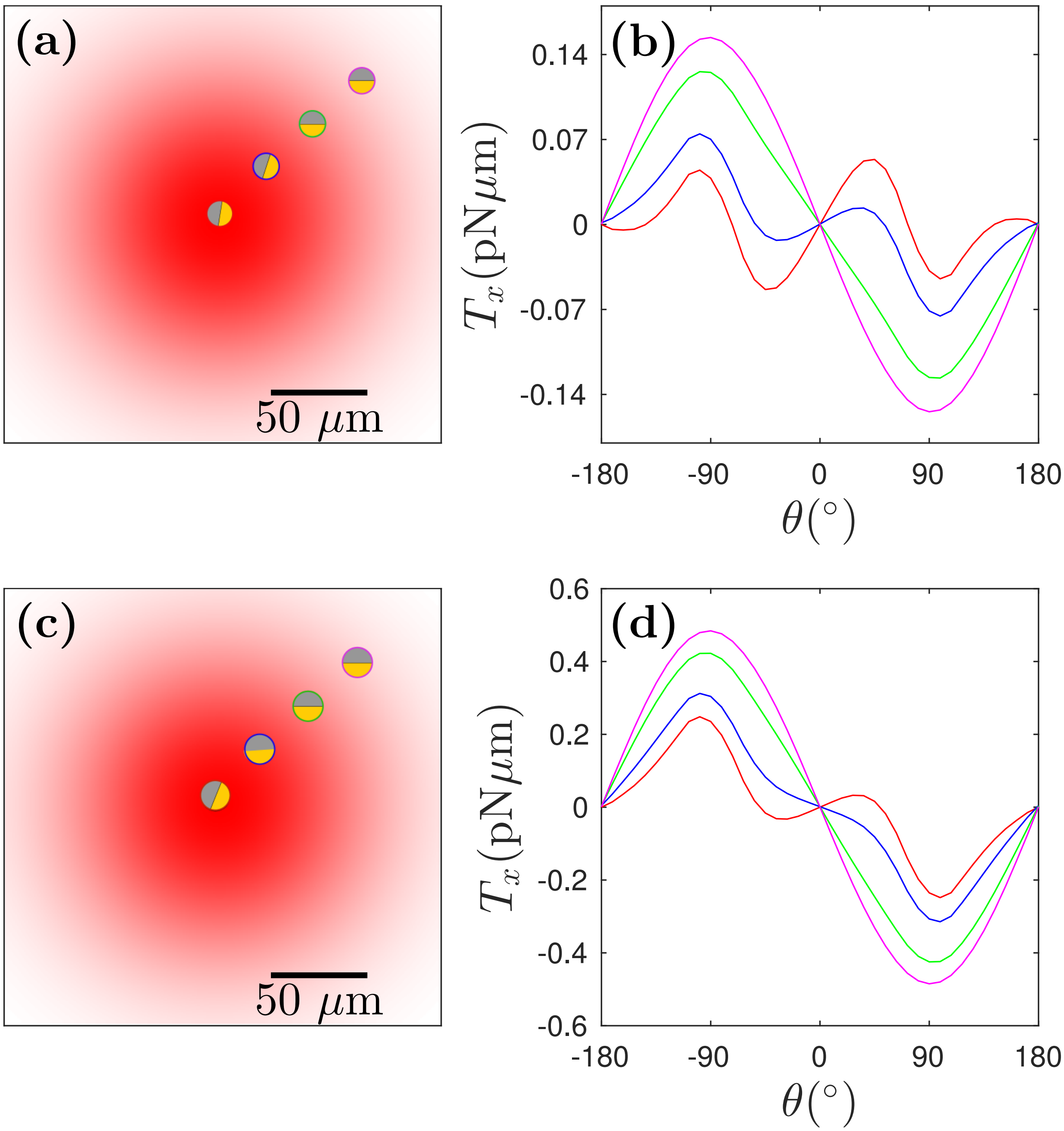


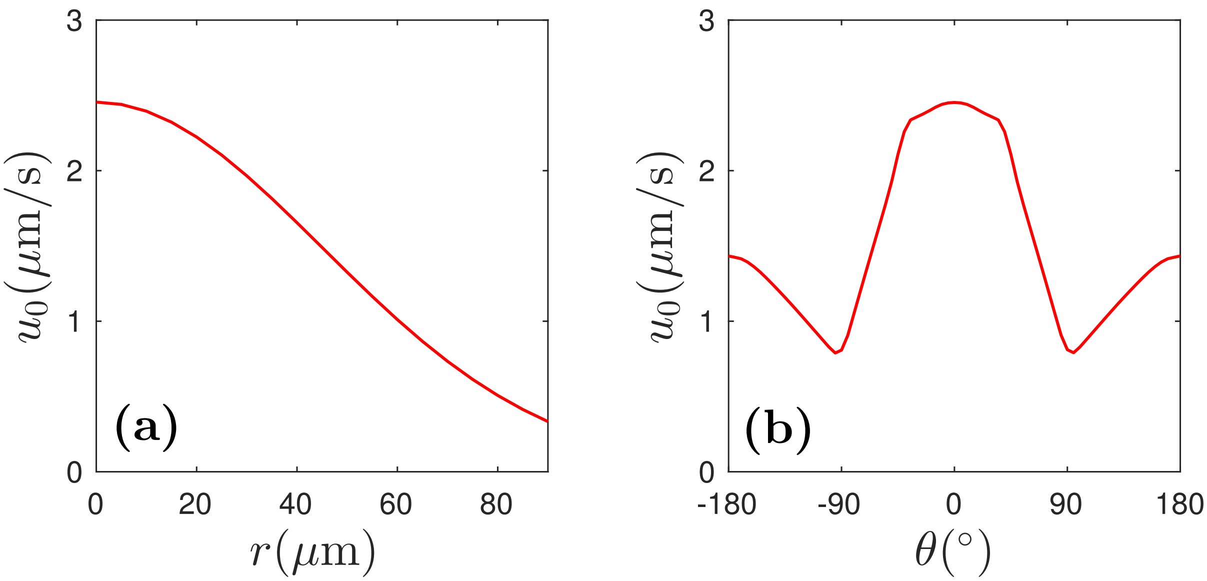
SUPPLEMENTARY FIG. 1: **Dependence of the optical force on the Janus particle's orientation angle.** The orientation of a Janus particle (silica, gold-coated, with 60nm gold layer thickness) is taken as fixed, and the optical force acting on the particle when set in a grid of points in the xy plane is calculated. (a) Optical force field in the xy-plane for a Janus particle when rotated clockwise of 20 degree around the x axis, starting from an initial orientation with cap down. The orientation of the particle with respect to the unit axes is sketched in (d) and (g). (b) Optical force field for a Janus particle with gold cap pointing downwards. The orientation of the particle with respect to the unit axes is sketched in (e) and (h). (c) Optical force field in the xy-plane for a Janus particle when rotated counterclockwise of 20 degree around the x axis, starting from an initial orientation with cap down. The orientation of the particle with respect to the unit axes is sketched in (d) and (g). The arrows length for each force field in (a,b,c) is normalised to the respective maximum magnitude of each case, in order to show clearly the direction of the force and its strong dependence on the orientation of the particle. The information about the magnitude is not shown here, but it is visible in Supplementary Fig. 2. The Gaussian potential is shown in a red colour scale proportional to its intensity. The size of the particle here is not specified because this behaviour happens for both 4.77 μm- and 6.73 μm-sized Janus particles.



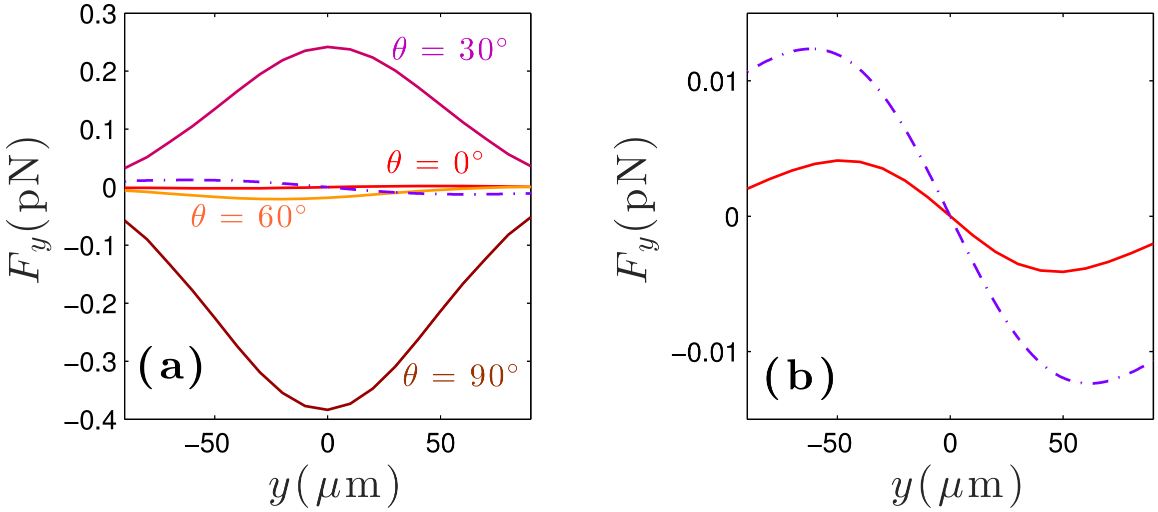
SUPPLEMENTARY FIG. 2: **Optical forces and torques for a 6.73 μm Janus particle**. (a) Schematic model of a Janus particle with its gold coated hemisphere oriented downwards. The direction of the principal axes is indicated. For a positive displacement along the y axis, the direction of the nonzero component of the optical force (F_y and F_z) is indicated. (b) Optical force (red, solid), thermophoretic force due to the absorption of optical intensity on the gold cap of a Janus particle on the particle itself (blue-green, dashed), thermophoretic force due to the beam (purple dashed-dotted line) along the direction of the y axis, as a function of the y-position of the center of the particle, when oriented like in panel (a). (c) Optical force (red, solid), thermophoretic force (blue-green, dashed) and vectorial sum of weight and buoyancy forces (dotted) along the z-axis, , when oriented like in panel (a). (d) A Janus particle with an orientation θ (considered rotating around the x axis). The Gaussian profile on top represent the beam intensity profile along the radial direction in the xy plane. The propagation direction is along the negative z axis. $\theta = 0$ corresponds to the orientation with cap down. (e) Optical force (solid-red), thermophoretic force (blue-green, dashed) and total force (solid-blue) as a function of rotation angle θ of the particle. (f) Optical torque (solid-red), buoyancy torque (dotted) and total torque (solid-blue) along the rotation axis (x-axis). From the trend shown in panel (e), we have that the optical force component F_y is in the direction of the line pointing from the silica half to the gold cap for small inclination angles θ, while it is in the opposite direction (from the gold cap to the silica half) for orientation angles θ close to 90°. Moreover, we see that the thermophoretic force is always in the direction of the gold cap, but the sum of the two components (optical plus thermophoretic) of the forces follows the trend of the optical force. From the trend shown in panel (f), we have that the total torque for the central position is such that the equilibrium orientation of the particle is not $\theta = 0$, i.e., with gold cap down, but it is some angle around $60 - 70°$. For this value of intensity, the torque due to the buoyancy is not able to stabilise the orientation of the particle towards maintaining the cap down.



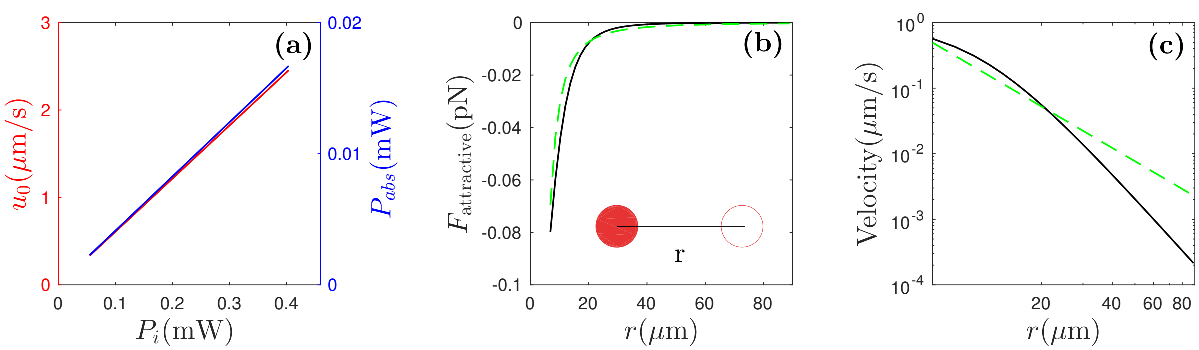
SUPPLEMENTARY FIG. 3: **Dependence of the optical torque on the position and the size of the Janus particle**. (a) Schematic model of a Janus particle at different distances from the beam center for a 4.77 μm Janus particle. (b) Total torque on a 4.77 μm Janus particle as a function of the orientation angle of the particle. (c) Schematic model of a Janus particle at different distances from the beam center for a 6.73 μm Janus particle. (d) Total torque on a 6.73 μm Janus particle as a function of the orientation angle of the particle. The torque is shown by (i) red lines when the particle is at the centre of the optical beam ($d = 0$), (ii) blue lines when the particle is at a distance $d = 35\,\mu$m from the centre, (iii) green lines when the particle is at a distance $d = 70\,\mu$m from the centre and by (iv) magenta when particle is at a distance $d = 105\,\mu$m. The incident beam has a beam waist $w_0 = 90\,\mu$m and a power $P = 100\,$mW.



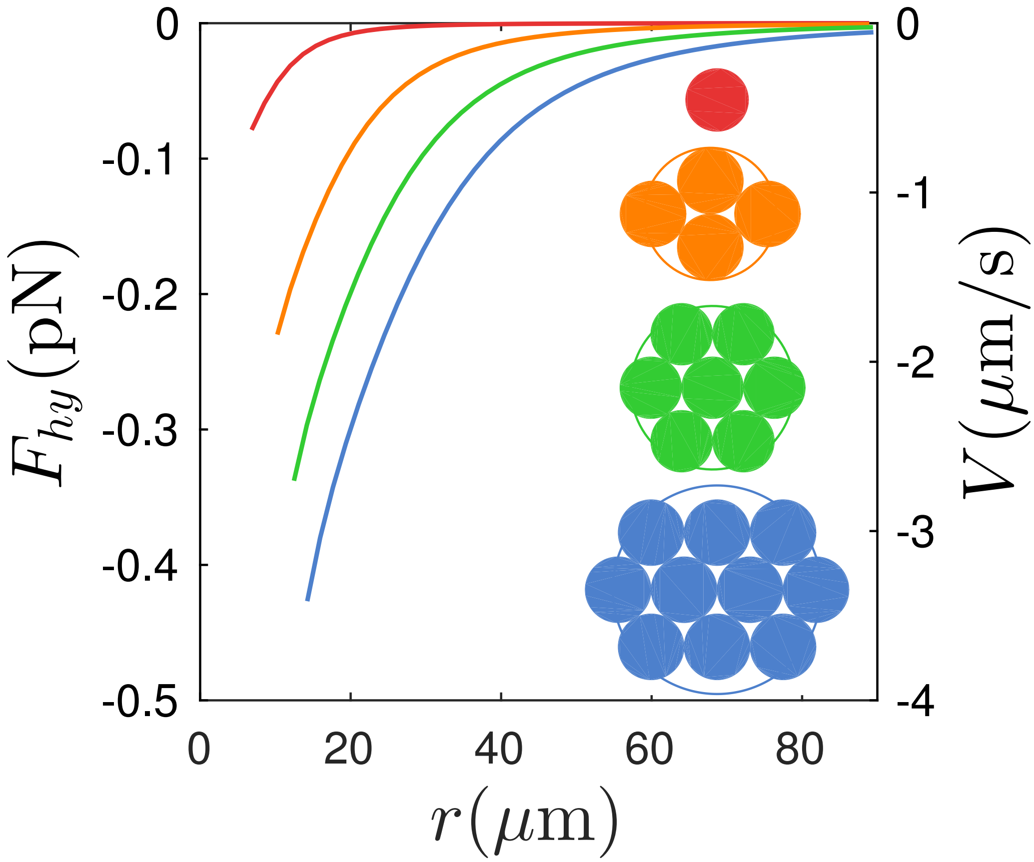
SUPPLEMENTARY FIG. 4: **Thermophoretic slip velocity of a Janus particle as a function of its distance from the beam center and of its orientation.** (a) Dependence of the thermophoretic slip velocity of a Janus particle versus its distance from the center of the optical beam. The gold cap of the Janus particle is oriented downwards. (b) Dependence of the thermophoretic slip velocity of a Janus particle versus its orientation. The values are calculated for a Janus particle of 6.73 μm size that is placed at the center of the beam. The thermophoretic velocity u_0 is as a function of the temperature gradient on the particle surface. As the dependence on the orientation θ shows, the minimum of the temperature gradient is for $\theta = 90°$, as expected. In such configuration, the power absorbed by the gold cap is minimum.



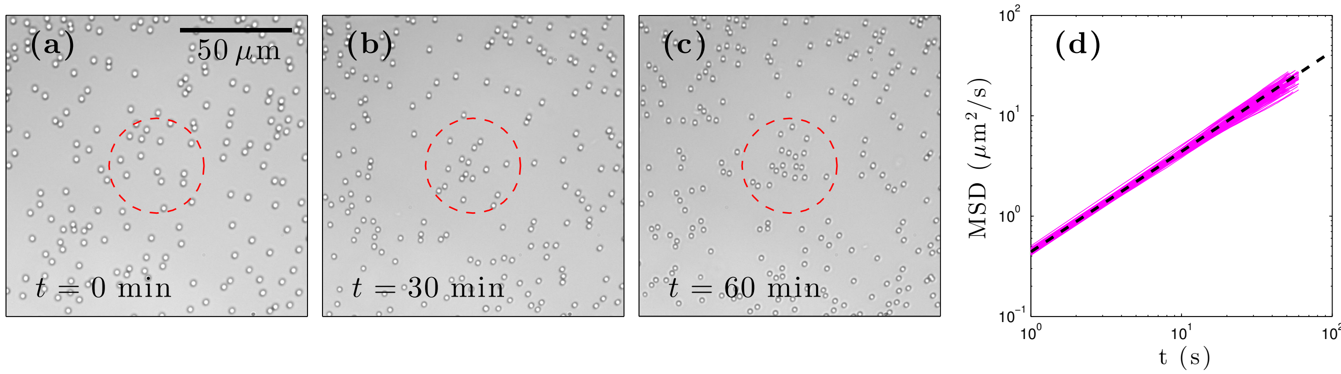
SUPPLEMENTARY FIG. 5: **Thermophoretic force of the beam compared to the optical force.**(a) Thermophoretic force (purple, dashed-dotted line) on a Janus particle (size 6.73 μm), compared with the optical force acting on a Janus particle (size 6.73 μm) for different orientations: $\theta = 0°, 30°, 60°, 90°$ (labels color-coded according to the respective lines). For the inclinations around $\theta = 30°$ and $\theta = 90°$ the optical force dominates on the thermophoretic force due to the beam. For inclinations around $\theta = 0°$ and $\theta = 60°$ the thermophoretic force due to the beam is of the same order of magnitude of the optical force. (b) Thermophoretic force (purple, dashed-dotted line) on a silica particle (size 6.73 μm) compared with the optical force (red line). For 6.73 μm-sized silica particles the optical force is comparable to the thermophoretic force due to the beam. The value of the thermophoretic mobility for a 6.73 μm-sized Janus particle and for 6.73 μm-sized silica particle is $D_T = -30\,(\mu\mathrm{m})^2\,\mathrm{s}^{-1}\,\mathrm{K}^{-1}$.



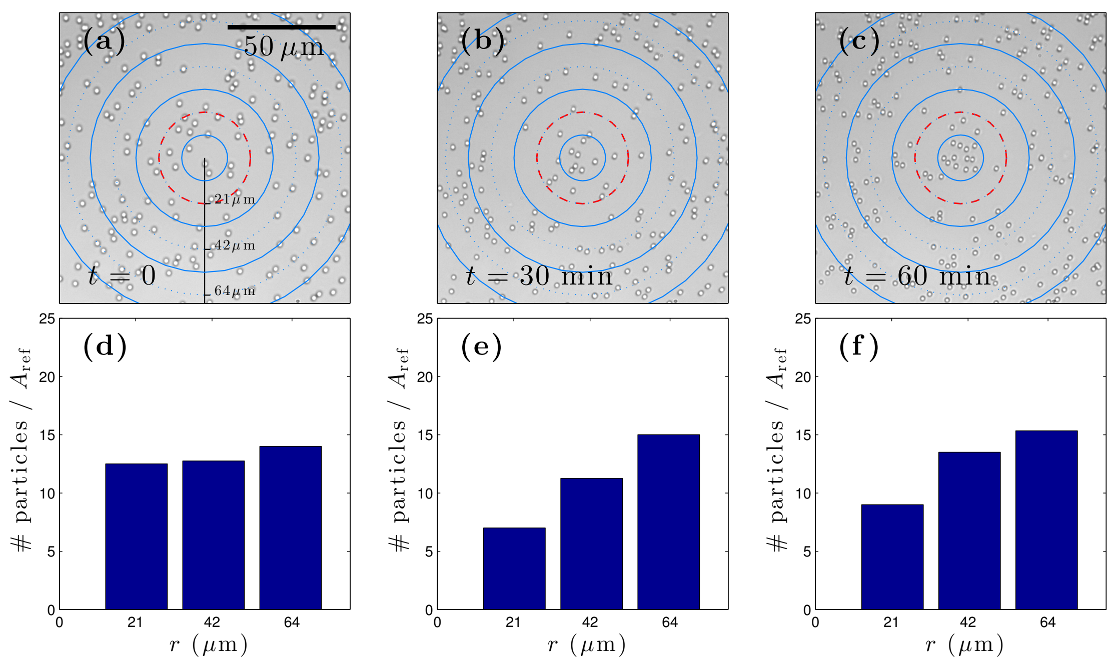
SUPPLEMENTARY FIG. 6: **Hydrodynamic attractive force induced by the presence of a hydrodynamic flow generated by a Janus particle, and thermophoretic force induced by a Janus particle**(a) Magnitude of the slip velocity and absorbed power as a function of the incident power for a Janus particle of 6.73 μm size. (b) Hydrodynamic force (black line) and thermophoretic force (green dashed line) in the proximity of a wall due to a 6.73 μm Janus particle as a function of the distance. (c) Hydrodynamic velocity (black line) and thermophoretic velocity (green dashed line) in the proximity of a wall due to a 6.73 μm Janus particle as a function of the distance. The value of the thermophoretic mobility for a 6.73 μm Janus particle is $D_T = -30\,(\mu\mathrm{m})^2\,\mathrm{s}^{-1}\,\mathrm{K}^{-1}$. The thermophoretic mobility of the particle subject to the thermophoretic interaction is also $D_T = -30\,(\mu\mathrm{m})^2\,\mathrm{s}^{-1}\,\mathrm{K}^{-1}$. The Janus particle is set at the center of a Gaussian beam (power $P = 100\,\mathrm{mW}$, beam waist $w_0 = 90\,\mu\mathrm{m}$).



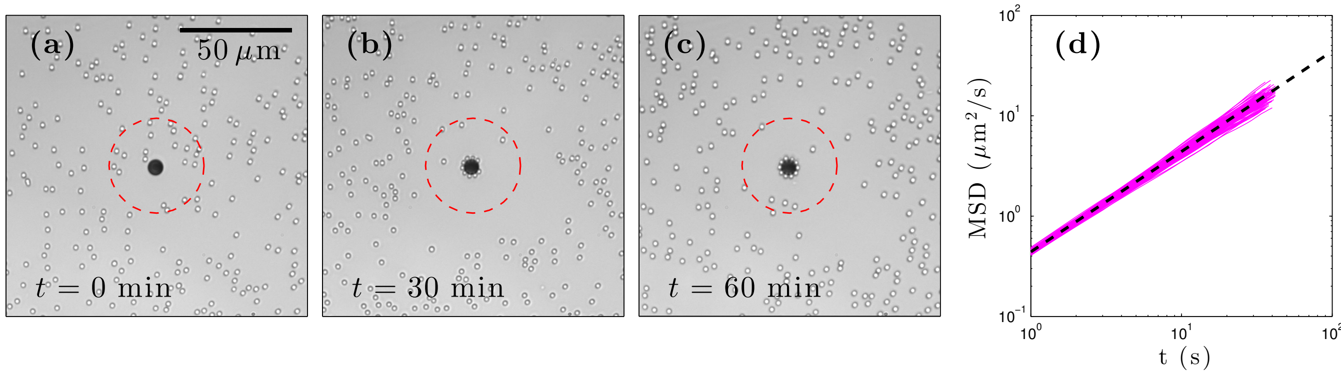
SUPPLEMENTARY FIG. 7: **Hydrodynamic force versus distance from the center of the cluster as a function of the cluster size.** The experimental observation is that a cluster of Janus particles generate a bigger hydrodynamic attractive force than a single particle Janus particle, and that also the action range of the attractive interaction of a cluster is bigger than the one of a single Janus particle. In order to model this behaviour, we assume that a cluster generates a hydrodynamic flow that is equivalent to the flow generated by a Janus particle with a radius that gives the same gold surface of the cluster. The radius of the cluster-equivalent Janus particle cluster is depicted by a circle centered in the geometrical center of the cluster. The diameter of a each single Janus particle in this case is 6.73 μm. The behaviour of the attractive force generated according to our assumption mimics the experimental observed behaviour. On the left, the scale indicates the attractive force produced by a cluster of Janus particles on a single 6.73 μm-size particle set at a distance r from the center of the cluster. The values given reflect the situation of a cluster located at the centre of the Gaussian beam. On the right, the corresponding flow velocity is indicated.



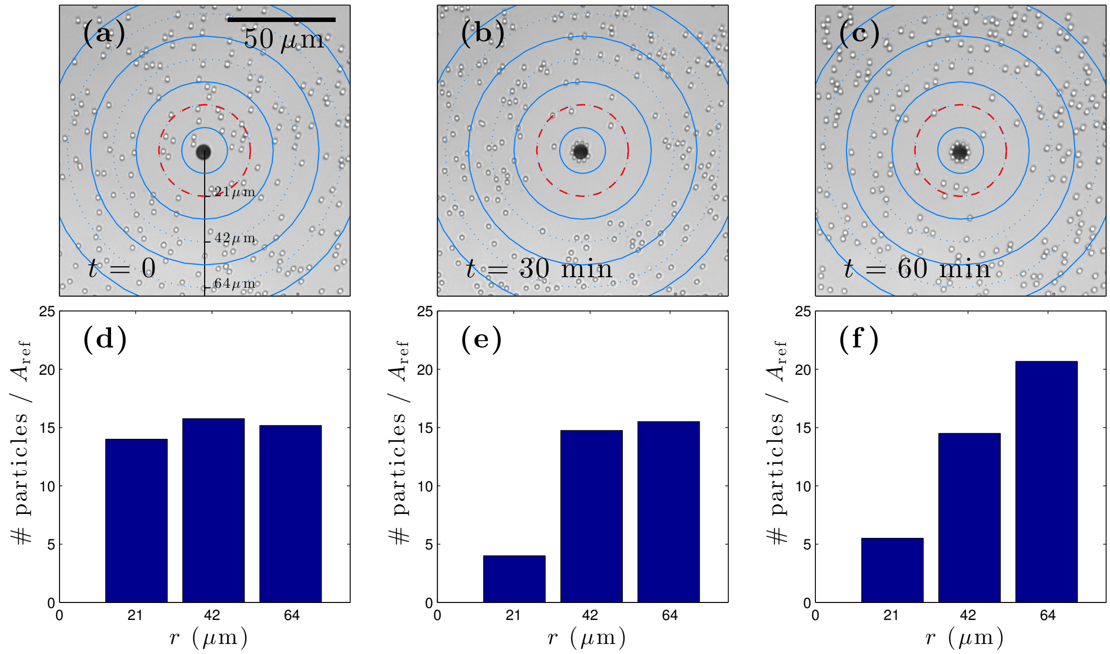
SUPPLEMENTARY FIG. 8: **Control experiment: only tracers** (a) initial, (b) mid-time and (c) final screenshot of the acquisition with a Gaussian optical potential of beam waist $w_0 = 30$ μm, power $P = 10$ mW (whose central spot is represented by the red dashed line), on a sample with tracer particles (silica, 2 μm size) only. The particles perform their Brownian motion, as one can see in the Supplementary Movie 10†, and only at the center of the screen there is a slight accumulation of particles that are attracted towards the center of the optical intensity. The optical force attracts the particles but allows some of them to escape. The particles that are farther than a radius of 30 μm are not experiencing any appreciable optical force, and their motion is purely Brownian. From the tracking of the particles we obtain the MSD, shown in (d). The black dashed line represents the MSD of an ideal Brownian particle of 2 μm size.



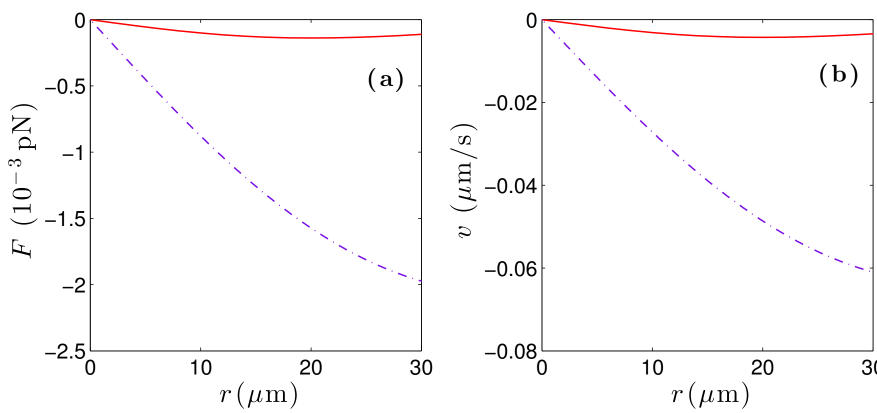
SUPPLEMENTARY FIG. 9: **Number of particles in a reference area for experiment with tracer particles only.** (a) initial, (b) mid-time and (c) final screenshot of the acquisition with a Gaussian optical potential of beam waist $w_0 = 30$ μm, power $P = 10$ mW (whose central spot is represented by the red dashed line), on a sample with tracer particles (silica, 2 μm size) only, with superposed the partition in concentrical circular regions. (d,e,f) Concentration of tracer particles in each of three non superposing circular areas, expressed as the number of particles found in a reference area Aref (indicated by the red circle) with the concentration equal to the concentration of each ring. (a,d) When the potential is switched on, the particles are equally concentrated in each of the three region, while as the time passes (b,e) and (c,f) the inner region shows a slight decrease in concentration, as the particles ends up in the central spot of the potential.



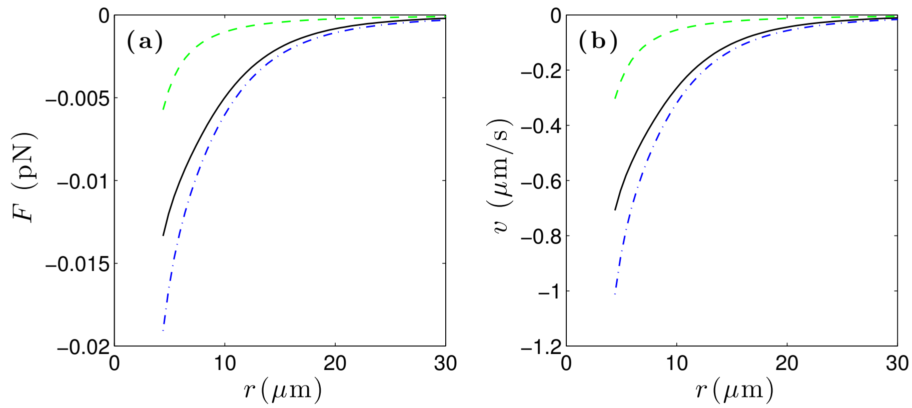
SUPPLEMENTARY FIG. 10: **Control experiment: stuck Janus particle and tracers.** (a) initial, (b) mid-time and (c) final screenshot of the acquisition with a Gaussian optical potential of beam waist $w_0 = 30$ μm, power $P = 10$ mW (whose central spot is represented by the red dashed line), on a sample with a stuck Janus particle set at the center of the optical intensity, with tracer particles (silica, 2 μm size). In this case, while the motion of the tracer particles far from the stuck Janus particle is mainly Brownian, the particles that in their erratic motion come close to the stuck Janus particle end up in getting drawn to the stuck particle. The arrangement, in a form of "flower" geometry, is reversible: the particles are still slightly moving close to the surface of the stuck Janus particle (See Supplementary Movie 11†). The MSD represented in (d) for the tracer particle in the region external to a 30 μm radius from the particle is similar to the one of the case with no Janus particle (the black dashed line represents the MSD of an ideal Brownian particle of 2 μm size). The particles that are instead closer to a distance of 10 μm from the Janus particle are attracted to it (See Supplementary Movie 11†)



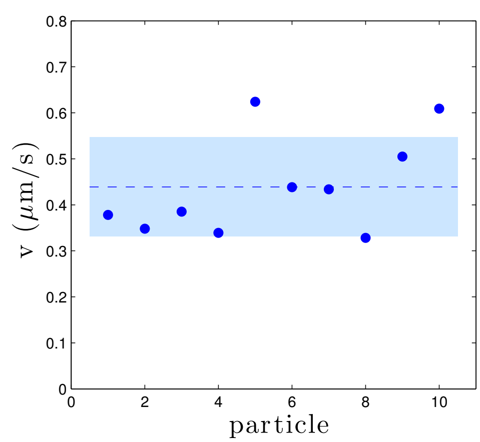
SUPPLEMENTARY FIG. 11: **Number of particles in a reference area for experiment with a stuck Janus particle and tracer particles.** (a) initial, (b) mid-time and (c) final screenshot of the acquisition with a Gaussian optical potential of beam waist $w_0 = 30$ μm, power $P = 10$ mW (whose central spot is represented by the red dashed line), on a sample with tracer particles (silica, 2 μm size) only, with superposed the partition in concentrical circular regions. (d,e,f) Concentration of tracer particles in each of three non superposing circular areas, expressed as the number of particles found in a reference area Aref (indicated by the red circle) with the concentration equal to the concentration of each ring. (a,d) When the potential is switched on, the particles are equally concentrated in each of the three region, while as the time passes (b,e) and (c,f) the inner region shows a slight decrease in concentration. The particles coming close enough to the Janus particle at the center remain attached to it.



SUPPLEMENTARY FIG. 12: **Control experiment: optical force, thermophoretic force due to the beam on a 2 μm silica particle, and corresponding velocities.** (a) Optical force (red) and thermophoretic force (purple dash-dotted line) due to the temperature increase induced in the sample from the Gaussian beam and velocity dependence for a tracer silica particle (2 μm size) in a Gaussian optical potential with $w_0 = 30$ μm and $P = 10$ mW. (b) optical velocity (red) and thermophoretic velocity (purple dash-dotted line) on a tracer silica particle (2 μm size) in a Gaussian optical potential with $w_0 = 30$ μm and $P = 10$ mW. The magnitude of both forces (and corresponding velocities) are negligible with respect to the hydrodynamic force and thermophoretic force due to the Janus particle (and corresponding velocities), as shown in Supplementary Fig. 13.



SUPPLEMENTARY FIG. 13: **Control experiment: hydrodynamic force and thermophoretic force due to the Janus particle on a 2 μm silica particle, and corresponding velocities.** (a) Force and (b) velocity as a function of radial distance for a tracer silica particle (2 μm size) for the hydrodynamic flow (black line) and the thermophoretic interaction (green dashed line) due to a Janus particle (6.73 μm size) stuck at the center of a Gaussian optical potential with $w_0 = 30$ μm and $P = 10$ mW. The blue dashed dotted line represents the (a) total force and (b) total velocity of the silica particle.



SUPPLEMENTARY FIG. 14: **Control experiment: experimental radial velocity of a 2 μm silica particle.** Experimental radial velocity in the last instants before touching the particle for each one of the ten particles attaching to the stuck Janus particle: the average radial velocity towards the Janus particle in proximity of the particle itself (within 8 μm to 4 μm from the center of the stuck Janus particle) is about 0.43 μm/s, in the range of the prediction of our model, as shown in Supplementary Fig. 13.