Supplementary Information for the paper "Active Cargo Transport with Janus Colloidal Shuttles using Electric and Magnetic Fields"

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Supplementary Notes

Estimation of the effective radius of the clusters

Shuttle-cargo clusters were experimentally found to move at constant velocity under the applied electric field. To predict the mean velocity of the self-propelling cluster, we assume the drag force, ^{*F*}_{Drag}, to be equal to the self-dielectrophoretic force that pushes the Janus particle, F_{sDEP} . $F_{sDEP} = F_{Drag}$. For a sphere, the drag force reads as $F_{Drag} = -6\pi\eta a\nu$, where v is the particle velocity, η is the viscosity of the liquid medium, and a is the radius of the particle. To apply this simple relation to the cluster comprising the Janus shuttle and a variable number of cargos, we assume an effective radius, a_{eff} , for the cluster of particles. The radius a_{eff} was estimated using two distinct approaches depending on the size of the cargo particle relative to the size of the Janus shuttle. For cargo particles smaller than the shuttle (2.85 µm), clusters were formed by one Janus colloid surrounded by several cargos (Figure 3a). In this case, the effective radius of the cluster was estimated by adding the cross-sectional areas of all colloids perpendicular to the velocity vector. The radius a_{eff} of a sphere displaying the same total cross-sectional area was used for the drag calculations. For the assembly where the cargo and Janus particles show comparable size (5.45 µm), the cargo and the shuttle move side-by-side perpendicular to the velocity vector (Figure 3b). This results in a dimer cluster whose drag force is better estimated by the addition of the individual drag forces, $F_{Drag-dimer} = -6\pi\eta(a_1 + a_2)v_{24}$, where a_1 and a_2 are the radii of the Janus and cargo particles. Thus, $a_{eff} = (a_1 + a_2)$ for this second cluster configuration. Figure 3c shown in the main text was generated based on these assumptions.

Speed of colloidal shuttles coated with metal layer

A balance between the dielectrophorectic and drag forces exerted on the colloidal shuttles indicates that the velocity of the colloidal shuttle (*v*) shows a quadratic dependence on the particle size, *a* (see main text): $v \sim a^2$. For a colloid shuttle of 4.64 µm and a typical metal coating of 15 nm, this scaling predicts a minor velocity increase of 0.65% if an extra layer of nickel is included on the surface of the shuttle.

Supplementary Figures



Supplementary Figure S1 | Estimation of self-propelling force that drives the motion of the Janus shuttle. FEA simulations of the electric field strength around the Janus colloid close to the electrode (Figure 2c, main text) allows for the estimation of the force that drives the shuttle particle. Note that the force is positive at distances between 4 and 8 μ m along the green line shown in Figure 2c in the main text.



Supplementary Figure S2 | Alignment of the nickel-coated Janus colloid under an external magnetic field. The sketch illustrates the differences between autonomous and magnetically-guided Janus spheres. The Janus colloid with only a platinum layer on one hemisphere moves autonomously independent of the applied magnetic field. By contrast, the Janus colloid with an additional nickel coating exhibits an induced magnetic dipole that favors the alignment along the

field direction. This enables magnetic steering of the self-propelling Janus spheres and allows for programming the delivery of cargo. Note that a uniform magnetic field is sufficient to magnetically guide the self-propelling particle, since the asymmetry (or magnetic dipole) needed for magnetic response is introduced in the particle itself. Experimentally, the static permanent magnet generates a field gradient. This gradient is not significant in our system because the magnet is placed at a distance of 4 cm away from the sample cell. At this large distance the field lines are relatively uniform over the area of our sample cell, which is approximately 18x18 mm.



Supplementary Figure S3 | Speed of a Janus shuttle plotted in In-In scale against the effective radius (aeff) of shuttle-cargo clusters with increasing number of cargos. The slope close to -1 of the obtained linear fits supports the scaling relations proposed in the main text.

Supplementary Movies

Movie S1 shows the trapping of 1 μ m spherical polystyrene particles around a Janus colloid and the autonomous motion of the resulting cluster when the electric field is on. Turning the field off releases the cargo and stops the motion. Applying an electric field again makes the shuttle recapture the cargos and continue its motion towards another random direction.

Movie S2 shows transfer of 1 μ m polystyrene cargos on the preprogrammed route shown in Figure 4a. At the final destination, the field is turned off to stop the motion of the shuttle-cargo cluster and to release the cargo.

Movie S3 depicts the pick-up, transfer and release of model 1 μ m polystyrene cargos on a preprogrammed zigzag (or stairs) route. Rotation of the magnet effectively changes the shuttle direction. At the final destination, the field is turned off to stop the motion of the cluster and to release the cargo.

Movie S4 demonstrates the programmed transport of 1 μ m polystyrene colloids on a house-shaped pre-designed route. During the path, the shuttle-cargo assembly moves along the diagonals twice. The numbers show the sequence of stretches that the shuttle went through as a function of time.

Movie S5 demonstrates the programmed transport of 1 μ m polystyrene colloids on a rectangular pre-designed route. When the assembly finishes the rectangular route it takes the diagonal path to conclude its tour.

Movie S6 demonstrates the transfer of $5.45 \,\mu m$ polystyrene colloids from the depot reservoir to the destination reservoir in a one-by-one sequential fashion as shown in Figure 5.