Electronic Supplementary Material (ESI) for NJC
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Supporting Information for
In-Situ Liquid Cell TEM Observation of Solution-Mediated Interaction Behaviour of Au/CdS Nanoclusters

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1 Some further ex situ characterizations on the CdS NCs and Au NPs sample

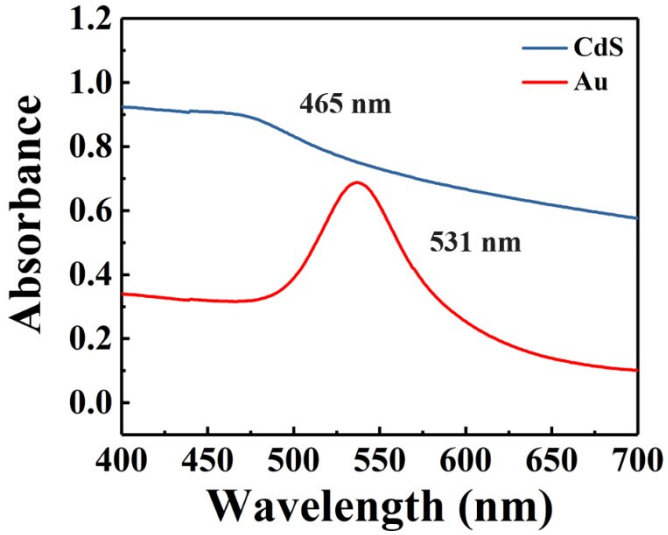

Fig. S1 The UV-Vis data of Au NPs and CdS NCs in the solution

The UV-Vis spectroscopies of the Au NPs and CdS NCs in aqueous solution sample are displayed in Fig. S1. The peaks of the ~60 nm Au NCs sample (red curve) and the ~5 nm CdS NCs sample (blue curve) are located at 531 nm and 465 nm, respectively, which are consistent with the literature [S3,S4]. The flat blue curve suggests that the CdS particles exist in agglomerated form with broad NC size distributions, while the single relatively narrow peak of red curve indicates the Au NPs were near monodispersed before the further treatment.

2. The relationship of liquid droplets' area and numbers

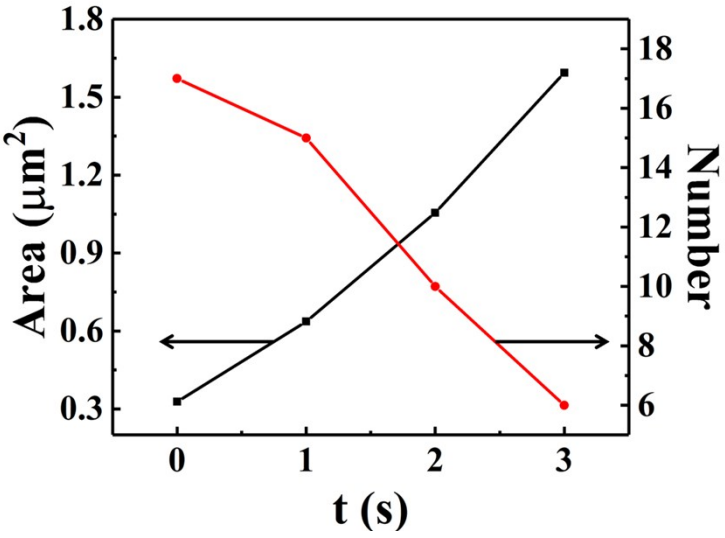

Fig. S2 The liquid droplets' total area and numbers vs. time curves.

3. The mean squared displacement (MSD) and velocity equation

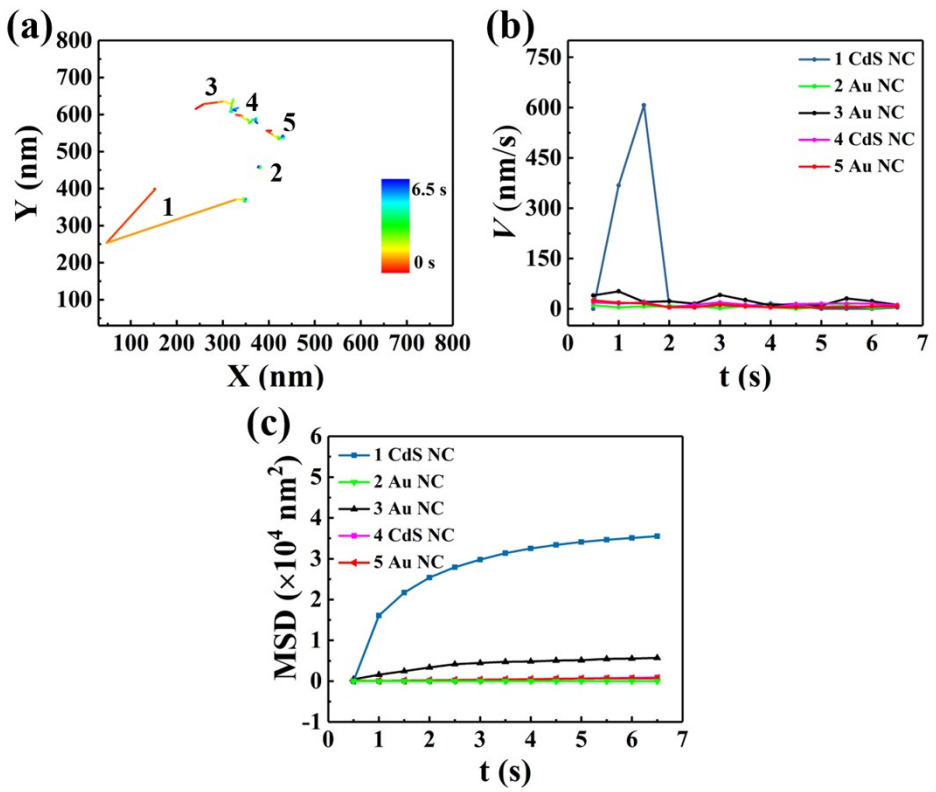

Fig. S3 Analysis of the NCs-1, 2, 3, 4 and 5 motion trajectories in Figure 3(b) and (c). (a) Trajectories of CdS and Au NCs. (b) *V* vs. *t* plot for the CdS and Au NCs. (c) The MSD $<r^2>$ of CdS and Au NCs as a function of time. The nonlinear curves of NC-1 indicate its D=MSD $<r^2>/4t$ values change with time. The ranges of D values are listed in Table 1 in the main text, in which, the minimum and maximum D values for NC-1 to 5 are estimated with the tangent properties of the data curves.

In statistical mechanics, the mean squared displacement (MSD) [S3, S4] is defined as

$$\mathrm{MSD} \equiv \mathrm{MSD} < r^2 >= \frac{1}{N}\sum_{n=1}^{N}\{[x_n(t)-x_n(0)]^2+[y_n(t)-y_n(0)]^2\} \qquad (1)$$

where N is the number of particles to be averaged, $x_n(0)=x_0$, $y_n(0)=y_0$, are the reference positions of each particle at time 0, $x_n(t)$, $y_n(t)$ are the positions of each particle at time t. The MSD of the five NPCs/NPs in our experiment in Fig. 3 in the main text are calculated and shown in Fig. S3.

The velocity (*V*) of the particles and the liquid front was calculated by the following equation [S5]

$$V = \frac{\sqrt{(x_{(t+\Delta t)}-x_{(t)})^2+(y_{(t+\Delta t)}-y_{(t)})^2}}{\Delta t} \qquad (1)$$

where $(x_{(t)}, y_{(t)})$ and $(x_{(t+\Delta t)}, y_{(t+\Delta t)})$ are the position coordinates of two adjacent data points (see in Fig. 2 and Fig. S3(a)), *t* is the time value of the video data, and Δt is the time interval.

The two-dimensional diffusion coefficients D of the NPs in Fig. 4 are further calculated using $D = <r^2>/4t$ [S6, S7], and the results are shown in Table S1. NC 1 was a small freely floating cluster before the interact event, and it shows an overall active diffusive motion trend. NC 2-5 were components in the large cluster, and are thus relatively inactive. NC 3 is located at the corner of the large cluster, and can perform some limited motions while keeping connected to the large cluster. The relative motions of the other components in the cluster are minimal.

Table S1 Two-dimensional diffusion coefficients of NCs in Fig. S3.

Nanoparticle Number	1	2	3	4		5
Time Range (s)	0-6.5	0-6.5	0-6.5	0-1.5	2-6.5	0-6.5
D/ (nm^2/s)	66-552	4-54	3-40	5263-8034	1682-4335	3-58

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