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Supplementary information

Photochromism from wavelength-selective colloidal phase segregation

In the format provided by the authors and unedited

1. **Supplementary Figures**

2. **Supplementary Tables**

1. Supplementary Figures

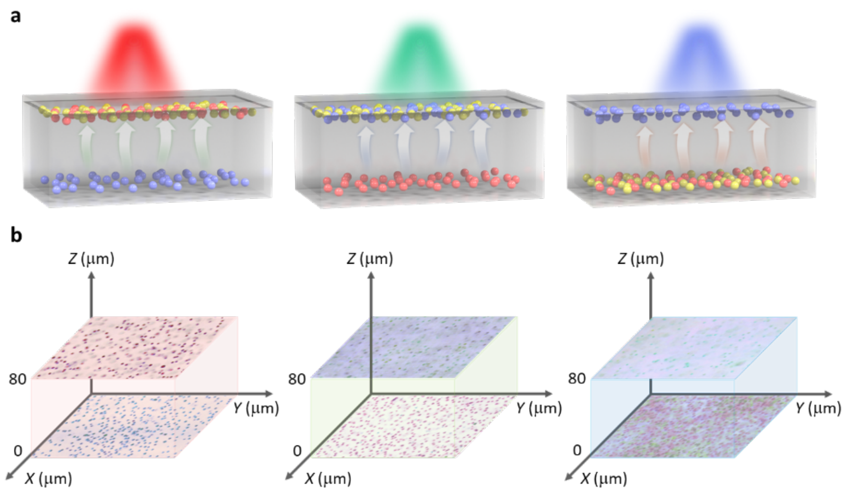

Fig. S1| Three-dimensional phase segregation of ternary colloidal system a, The schematic of three-dimensional phase segregation of the ternary colloidal system. A mixture solution of L0, LEG4, and SQ2 sensitized TiO_2 colloids is transferred into a transparent chamber. Different light illumination induces various vertical segregation. **b,** 3D distribution of the coloured colloids at different depths (Z=0, 80 μm) under red, green, and blue light illumination, respectively. L0 and LEG4 sensitized TiO_2, SQ2, and L0 sensitized TiO_2, SQ2 sensitized TiO_2 are pushed up to the top under red, green, and blue light illumination, respectively.

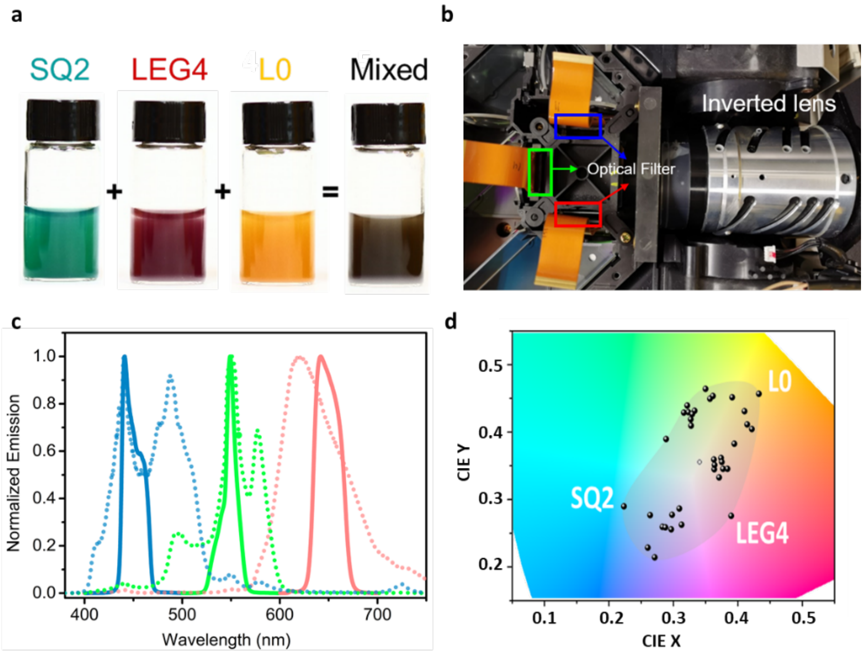

Fig. S2| Spectra-selective active colloidal mixture as photochromic colloidal swarm. a, The photograph of SQ2, LEG4, and L0 sensitized TiO_2 colloids and their mixed suspension, which appear as cyan, magenta, yellow and black ink, respectively. **b,** Modified 3LCD projector with optical filters. **c,** The normalized spectrum of the projector output for blue, green, and red light with (solid line) or without (dash line) optical filter. **d,** Experimental colour gamut of the photochromic colloidal swarm presented in a standard CIE-1931 colour space.

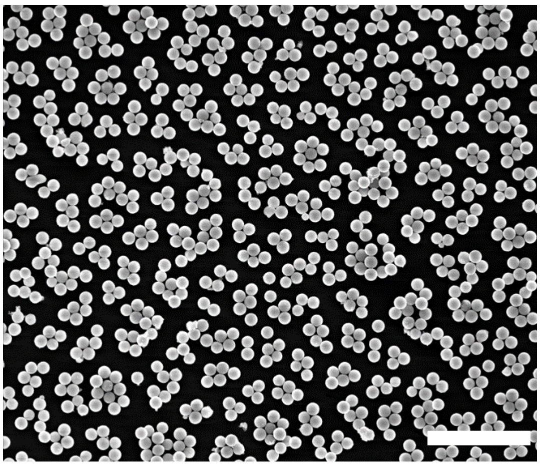

Fig. S3| The scanning electron microscopy (SEM) image of monodispersed TiO_2 microbeads. Scale bars: 10 μm.

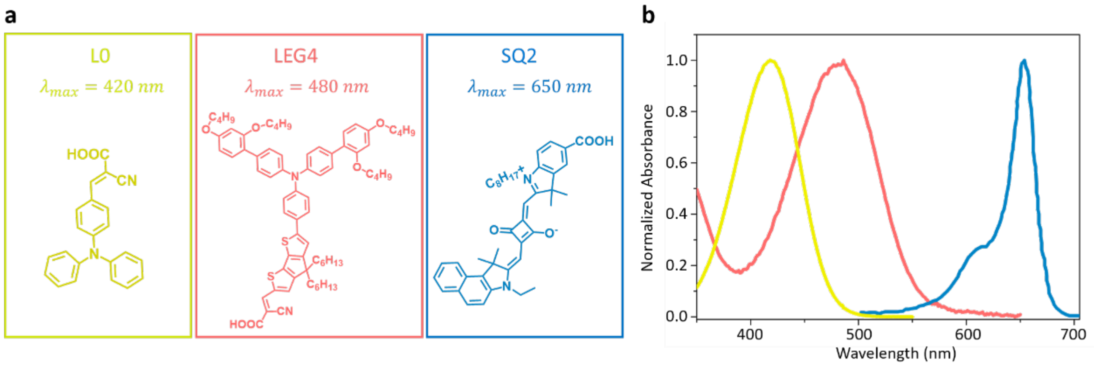

Fig. S4| Molecular structure and absorption spectrum of organic dyes a, The molecular structure and maximum absorption wavelength of dye L0, LEG4, and SQ2. **b,** The normalized absorbance of ethanolic solution of dye (L0 (yellow), LEG4 (magenta), and SQ2 (cyan)), with distinctive absorbance covering the visible spectrum.

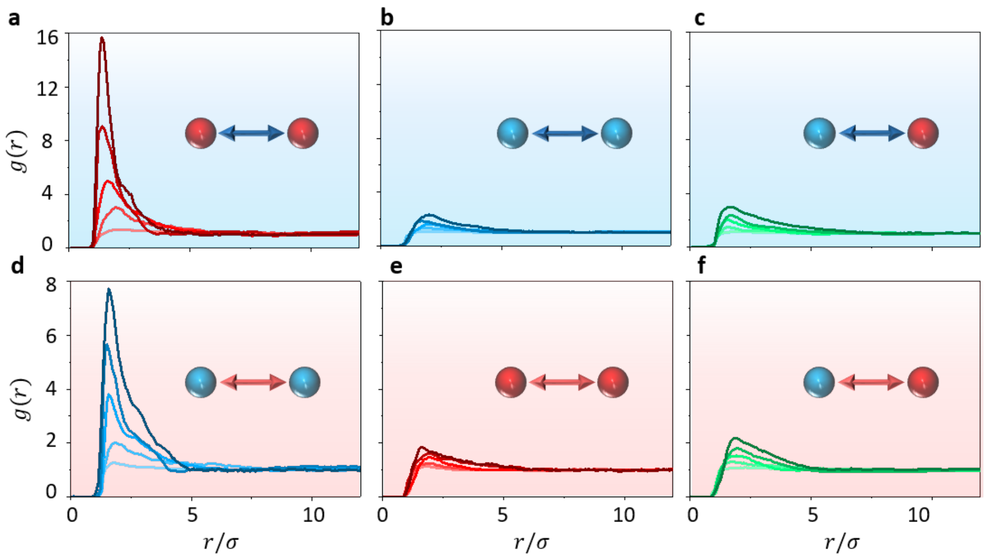

Fig. S5| Radial distribution function of dye-sensitized-colloid modulated with illumination. a-c, The $g(r)$ as a function of r/σ for LEG4-LEG4 sensitized TiO_2 colloids (**a**), SQ2-SQ2 sensitized TiO_2 colloids (**b**), and LEG4-SQ2 sensitized TiO_2 colloids (**c**) under blue light (440-480 nm) illumination with various intensity (10 mW/cm^2, 30 mW/cm^2, 50 mW/cm^2, 70 mW/cm^2, 90 mW/cm^2). **d-f,** The $g(r)$ for SQ2-SQ2 sensitized TiO_2 colloids (**d**), LEG4-LEG4 sensitized TiO_2 colloids (**e**), and LEG4-SQ2 sensitized TiO_2 colloids (**f**) under red light (640-660 nm) illumination with various intensity (10 mW/cm^2, 30 mW/cm^2, 50 mW/cm^2, 70 mW/cm^2, 90 mW/cm^2).

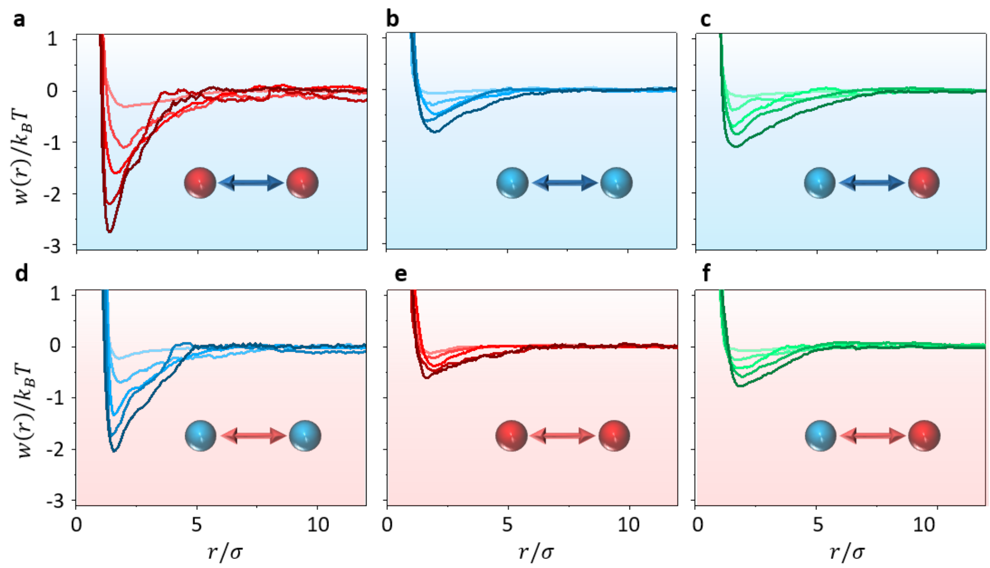

Fig. S6| Apparent pair potential of dye-sensitized-colloid modulated with illumination. a-c, The $w(r)$ as a function of r/σ for LEG4-LEG4 sensitized TiO_2 colloids (**a**), SQ2-SQ2 sensitized TiO_2 colloids (**b**), and LEG4-SQ2 sensitized TiO_2 colloids (**c**) under blue light (440-480 nm) illumination with various intensity (10 mW/cm^2, 30 mW/cm^2, 50 mW/cm^2, 70 mW/cm^2, 90 mW/cm^2). **d-f,** The $w(r)$ for SQ2-SQ2 sensitized TiO_2 colloids (**d**), LEG4-LEG4 sensitized TiO_2 colloids (**e**), and LEG4-SQ2 sensitized TiO_2 colloids (**f**) under red light (640-660 nm) illumination with various intensity (10 mW/cm^2, 30 mW/cm^2, 50 mW/cm^2, 70 mW/cm^2, 90 mW/cm^2).

2. Supplementary Tables:

Table S1. Fitting parameters under various blue light illumination

passive-passive	10 mW/cm^2	30 mW/cm^2	50 mW/cm^2	70 mW/cm^2	90 mW/cm^2
D_0 (k_BT)	0.1086	0.3643	0.5357	0.6894	0.8786
α (μm^{-1})	2.6526	1.7027	1.2636	1.3329	0.9301
r (μm)	1.5975	1.6775	1.9440	1.8631	2.0373
active-active	**10 mW/cm^2**	**30 mW/cm^2**	**50 mW/cm^2**	**70 mW/cm^2**	**90 mW/cm^2**
D_0 (k_BT)	0.4000	1.2503	1.7840	2.2361	2.7269
α (μm^{-1})	1.6438	1.1023	1.1132	1.4569	1.3855
r (μm)	1.7563	1.9024	1.7306	1.5977	1.5193
passive-active	**10 mW/cm^2**	**30 mW/cm^2**	**50 mW/cm^2**	**70 mW/cm^2**	**90 mW/cm^2**
D_0 (k_BT)	0.2084	0.4070	0.6449	0.8762	1.2472
α (μm^{-1})	2.1335	1.1827	1.1901	1.0952	0.9000
r (μm)	1.6116	1.9551	1.7884	1.8615	1.9441

Table S2. Fitting parameters under various red light illumination

passive-passive	10 mW/cm^2	30 mW/cm^2	50 mW/cm^2	70 mW/cm^2	90 mW/cm^2
D_0 (k_BT)	0.1212	0.2268	0.3492	0.5339	0.6908
α (μm^{-1})	2.6771	1.6444	1.3447	1.1297	1.2802
r (μm)	1.6707	1.7449	1.9612	1.8925	1.8015
active-active	**10 mW/cm^2**	**30 mW/cm^2**	**50 mW/cm^2**	**70 mW/cm^2**	**90 mW/cm^2**
D_0 (k_BT)	0.3016	0.7787	1.2406	1.6579	2.0594
α (μm^{-1})	1.2958	1.1339	1.1692	1.4006	1.1553
r (μm)	2.2428	2.0770	1.9855	1.8078	1.7847
passive-active	**10 mW/cm^2**	**30 mW/cm^2**	**50 mW/cm^2**	**70 mW/cm^2**	**90 mW/cm^2**
D_0 (k_BT)	0.1184	0.3530	0.4935	0.6431	0.8430
α (μm^{-1})	2.3619	1.7563	1.5617	1.2230	1.0367
r (μm)	1.6193	1.7640	1.7081	1.8746	2.0277

Table S3. Expected layered phase behaviour under different illumination spectra

Illuminated light	Light component	Top layer (passive)	Bottom layer (active)
Red	Red	L0-TiO_2 + LEG4-TiO_2	SQ2-TiO_2
Green	Green	L0-TiO_2 + SQ2-TiO_2	LEG4-TiO_2
Blue	Blue	SQ2-TiO_2	L0-TiO_2 + LEG4-TiO_2
Cyan	Green + Blue	SQ2-TiO_2	L0-TiO_2 + LEG4-TiO_2
Magenta	Red + Blue	LEG4-TiO_2 (weak active)	L0-TiO_2 + SQ2-TiO_2
Yellow	Red + Green	L0-TiO_2	LEG4-TiO_2 + SQ2-TiO_2

Table S4. The zeta potential of pristine TiO_2 and different dye-loaded TiO_2

Zeta Potential (mV)				
	pristine	L0-TiO_2	LEG4-TiO_2	SQ2-TiO_2
(+)TiO_2	+10.33	+8.23	+7.78	+8.72
(-)TiO_2	-38.45	-42.38	-48.68	-15.01

Table S5. Comparison of current state-of-the-art photochromic materials.

Type of Materials	Excitation Light	Response Time	Display Color	Mechanism	Ref.
Photochromic Colloidal Swarm	Full Spectrum (~10-20 mW/cm^2)	1 min	Full Spectrum (positive and negative response) No Erase needed	Phase Segregation	This Paper
Phosphomolybdic Acid	365 nm UV Flashlight (5 W)	5 min	Dark Blue Air oxidation erase	Valence Transition	[55]
Poly(ionic liquids) and Polyoxometalates	Projector Lamp (210 W)	2 min	Light Yellow and Bluish Green Air oxidation erase	Valence Transition	[56]
Bismuth Oxyhalide	365 nm UV (6 mW/cm^2)	1 min	Brown Black Air oxidation erase	Valence Transition	[57]
WO_3 Nanoparticles	UV lamp (5 W)	3 min	Blue Heat erase	Valence Transition	[58]
Photochromic Diarylethene Derivatives	254 nm UV (1 mW/cm^2)	30 min	Cyan or Magenta or Yellow Visible light erase	Isomerization	[59]
Photochromic Dyes	Full Spectrum (1200 Lumens)	20-120 min	Full Spectrum (positive response) UV (4W) erase	Isomerization	[60]