1	Electronic supporting information (ESI)
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3	Positive and negative birefringence in packed films of binary spherical colloidal particles
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4 Fig. S1 Optical micrographs of particulate films of mono-modal colloidal silica particles obtained by 5 drying of suspensions. The particle diameters were (a), (g), and (m) 5 nm, (b), (h), and (n) 12 nm, (c), (i), 6 and (o) 20 nm, (d), (j), and (p) 45 nm, (e), (k), and (q) 100 nm, and (f), (l), and (r) 200 nm. We observed 7 (a)–(f) in transmission mode, (g)–(l) under crossed polarizers, and (m)–(r) under crossed polarizers with 8 a compensator. All of the scale bars are 500 μ m. Clear birefringence was not observed in the films of 5 9 (m) or 12 nm (n). The film of 200 nm particles was rather turbid (r), probably because of light scattering 10 by the large particles.

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Fig. S2 SEM images of packed films of 100 and 12 nm particles with $\varphi_L = 0.7$, taken from different positions. (a) The thickest position and (b) Near the center of the sample. Scale bars are 1 µm. (c) shows a schematic illustration of a packed film of 100 and 12 nm with $\varphi_L = 0.7$, in which both positions for (a) and (b) are indicated.

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