Supporting Information

Formation of Two-Dimensional Diamond-Like Colloidal Crystals Using Layer-by-Layer Electrostatic Self-Assembly

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1. The refractive index of titania

Dielectric dispersion representing titania in the FDTD simulation was produced by fitting experimental data of dielectric dispersion of titania[S1] using Lorentz–Drude function based on quantum annealing. The dielectric dispersion of the Lorentz-Drude model, $\varepsilon^*(\omega)$ is given by

$$\varepsilon^*(\omega) = \varepsilon_0 \left\{ \varepsilon_\infty + \frac{f_d \omega_p^2}{-\omega^2 + j\omega\Gamma_d} + \sum_{n=1}^{m} \frac{f_n \omega_p^2}{\omega_n^2 - \omega^2 + j\omega\Gamma_n} \right\} . \quad \text{(SI1)}$$

Here, ε_0 is the permittivity of a vacuum, ε_∞ the relative instantaneous permittivity, ω_p the plasma frequency, Γ_d the damping frequency of the Drude term, f_d the oscillator strength of the Drude term, m the number of Lorentz terms, ω_n the resonance frequency of the *n*-th Lorentz term, Γ_n the damping frequency of the *n*-th Lorentz term, f_n the oscillator strength of the *n*-th Lorentz term, and ω the angular frequency of light. Table S1 shows the parameters of the Lorentz-Drude function.

Table S1. Parameters of Lorentz - Drude model determined by fitting the experimentally obtained dielectric function of titania.

m	1	
ε_∞	1	
ω_p	661.4411	THz
ω_1	1191.053	THz
Γ_d	0	THz
Γ_1	0	THz
f_d	1.1108×10^{-5}	
f_1	19.916	

2. FDTD simulation

Numerical EM-field analysis based on the FDTD method was performed by the commercial software Opti FDTD (Optiwave Systems Inc., Ottawa, Canada). Here, diamond structures of titania particles with 3- to 16-layers of 2D crystals and the refractive index (see SI1) in air were assumed. The *x*- and *y*- dimensions of the simulation box were fixed at 2.5 μm and 2 μm, respectively, and *z*- dimension was set to 1 μm (for the number of layers, N = 3 and 4), 1.5 μm (N = 8) and 2.5 μm (N = 16); from -1.25 to +1.25 μm in the *x*-coordinate, from 0 to +2 μm in the *y*-coordinate, and from 0 to a maximum of +2.5 μm in the *z*-coordinate). Anisotropic perfectly matched layer boundary condition was adopted for all the outer surfaces of the simulation box. A two-dimensional pulsed light source was set at z = 0.2 μm.

The waveform type, amplitude, center wavelength of the pulse wave, FWHM of the pulse and incident direction were set to Sine-Modulated Gaussian Pulse, 1.0 V/m, 700 nm (428.275 THz), 220.636 THz and +*z* to analyze the spectrum in the range from 500 nm to 1000 nm. The discretization conditions of spatial coordination and time domain were $\Delta x = \Delta y = \Delta z$ = 0.005 μm and $\Delta t = 9.244 \times 10^{-18}$ sec. Calculation for each structure was carried out for 30000 steps. Figure 10(c) of the main text shows the reflection and transmission spectra obtained from the numerical analyses.

Reference

S1 J. R. DeVore, *J. Opt. Soc. Am.*, 1951, **41**, 416-419.