Electronic Supplementary Material (ESI) for CrystEngComm.

Electronic Supporting Information (ESI) for

Enantioselective Amplification on Circularly Polarized Laser-Induced Chiral Nucleation from a $NaClO_3$ Solution Containing Ag Nanoparticles

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ESI1. Solution preparation

A supersaturated aqueous solution of $NaClO_3$ was prepared at 22°C by dissolving $NaClO_3$ powder (30 g, >98 %, Wako) to ultrapure water (25 mL), which is fabricated with Direct-Q 3UV (Millipore), in a 100 ml glass beaker. The solution was then heated up to 50°C while being stirred using hotplate magnetic stirrer to dissolve the $NaClO_3$ powder completely. After assuring the complete dissolution of the $NaClO_3$ powder, the resulting solution was transferred to a 25 mL centrifuge tube with a screw cap, and then hermetically closed. The centrifuge tube was left for a week at 22°C to precipitate the solute dissolving excessively, leading the solution to equilibrium state at 22°C. The equilibrium solution is the mixture of the $NaClO_3$ saturated aqueous solution and the $NaClO_3$ crystalline powder. The mixture was then treated in a centrifuge to separate the saturated solution from the crystalline powder. The supernatant of the mixture was used as a $NaClO_3$ aqueous solution saturated at 22°C.

ESI2. The degree of depolarization of CPL caused by the oblique reflection by the Notch-dichroic half mirror.

Figure ESI2 shows the optical system for the evaluation of the degree of depolarization caused by the oblique reflection by the Notch-dichroic half mirror. We evaluated the degree of the depolarization by comparing the degree of linear polarization of the light incident to the dichroic half mirror [Figure ESI2(a)] and that of the light converted from circularly polarized light after oblique reflection to elliptically polarized light by a quarter wave plate [Figure ESI2 (b)]. The degree of the linear polarization was evaluated by comparing intensity of s-polarization component and p-polarization component of the light. The polarization components was divided using polarization beam splitter and the intensity was measured using a power meter.

The measurement in (a) configuration shows that the intensity of s-component was 564 mW and p-component was 0 mW. On the other hand, the measurement in (b) configuration provided that s-component was 489 mW and p-component was 12 mW, indicating that the degree of linear polarization of the light incident to the dichroic mirror was 100% and that of the light depolarized by the mirror was 95.2%. The difference of the degree of the linear polarizations is 4.8%. Namely, assuming that a pure circularly polarized light was introduced to the inverted optical microscope we used, about 95% circular polarization is guaranteed before focusing by an objective lens.

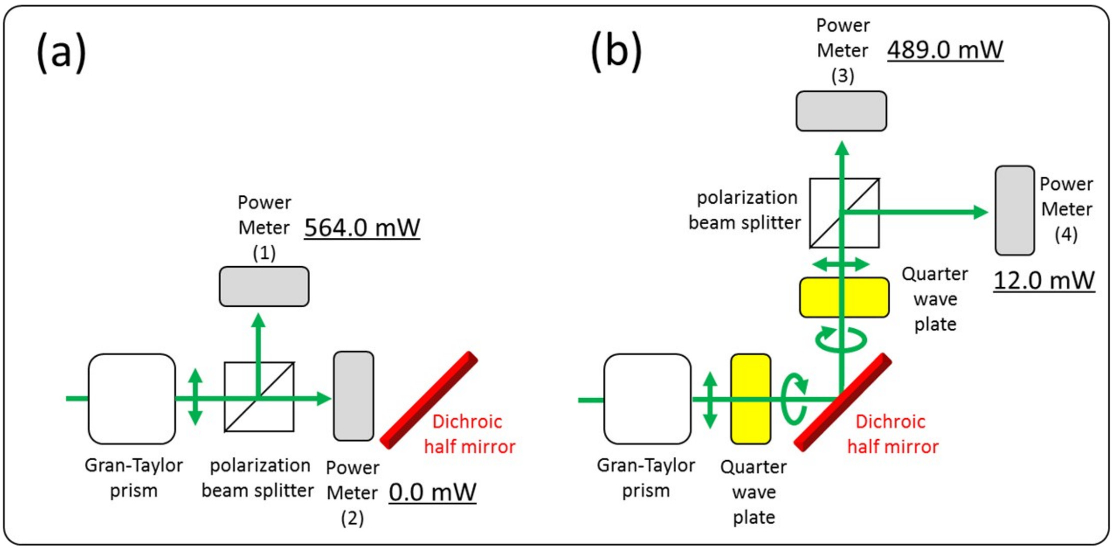

Figure ESI2. Schematics showing optical setups to evaluate the degree of depolarization by the oblique reflection by the dichroic half mirror.

ESI3. The Evaluation of the Ellipticity of the Circularly Polarized Light

The degree of the circular polarization of the light reflected by the Notch-dichroic half mirror set before the objective lens, ϵ, was evaluated by rotating analyzer method in accordance with following definition:

$$\epsilon = \sqrt{\frac{I_{min}}{I_{max}}} \qquad \text{(ESI3-1)}$$

where I_{min} is the light intensity in the minor axis of the elliptically-polarized light and I_{max} is the light intensity in the major axis of the light. ϵ can be a value from 0 to 1; the value of 1 indicates complete CPL and the value of 0 indicates complete linearly polarized light. Only the light whose ϵ is more than 0.93 was used as CPL in the current experiment.

ESI4. Movie of $NaClO_3$ crystallization process of the current laser-induced crystallization experiment.

Movie of *in-situ* polarized-light microscopic observation of $NaClO_3$ crystallization process from the focal spot induced by circularly polarized light laser trapping of Ag nanoparticles at the air/solution interface. The movie corresponds to Figure 2 a. The configuration of polarized light microscope is not complete crossed-Nicols but the angle of the analyzer was slightly rotated (a few degrees) from the crossed-Nicols configuration, leading to the bright microscopic view.

Figure ESI4. A captured micrograph of the in-situ polarized microscopic observation of $NaClO_3$ crystallization process from the focal spot induced by circularly polarized light laser trapping of Ag nanoparticles at the air/solution interface.

ESI5. Experimental procedure of circular dichroism measurement (Figure ESI5)

Single crystals of *d*- and *l*-chiral cubic phase with dimensions of 5mm×5mm×2.5mm were grown by evaporating a saturated $NaClO_3$ aqueous solution (2.5 mL) in a 5 mL container. The grown crystal was transparent and faceted to (100) faces. The grown crystal was set on a sample stage which is specially-designed for J-1100 (Jasco, Japan), circular dichroism spectrometer. The specially-designed stage was designed so that a specimen can be rotated around the axis of the incident light, allowing us to measure the dependence of CD on crystal orientation. The crystal was set on the stage so that the light incidence becomes perpendicular to a (100) face constructed by the side of 5mm and the side of 2.5mm, namely, the length that the light passes through the crystal is approximately 5 mm. CD spectra from 200 nm to 600 nm were recorded with the wavelength resolution of 0.1 nm for each measurement. The measurement was repeated every 45^{o} rotation of the crystal orientation from 0^{o} to 360^{o}. The same set of the procedure was applied also for the CD measurement of the opposite enantiomorph. After the CD measurement, Ag thin films were deposited on the incidence (100) face and the opposite $(\bar{1}00)$ face of the sample crystals by a resistant heat vacuum vapor deposition method using VE-2012 (Shinku Device Co., Ltd., Japan). Ag thin film was grown by evaporating a silver wire (ϕ = 0.2, 4 mm length) on a Tungsten basket applied a 20 A current under pressure of 3.6×10^{-4} Pa. The sample crystal with Ag thin film exhibited purple semi-transparent color, resulting from light absorption by surface plasmon resonance of the Ag thin film. The procedure for CD measurement written above was applied to the sample crystals with Ag thin film.

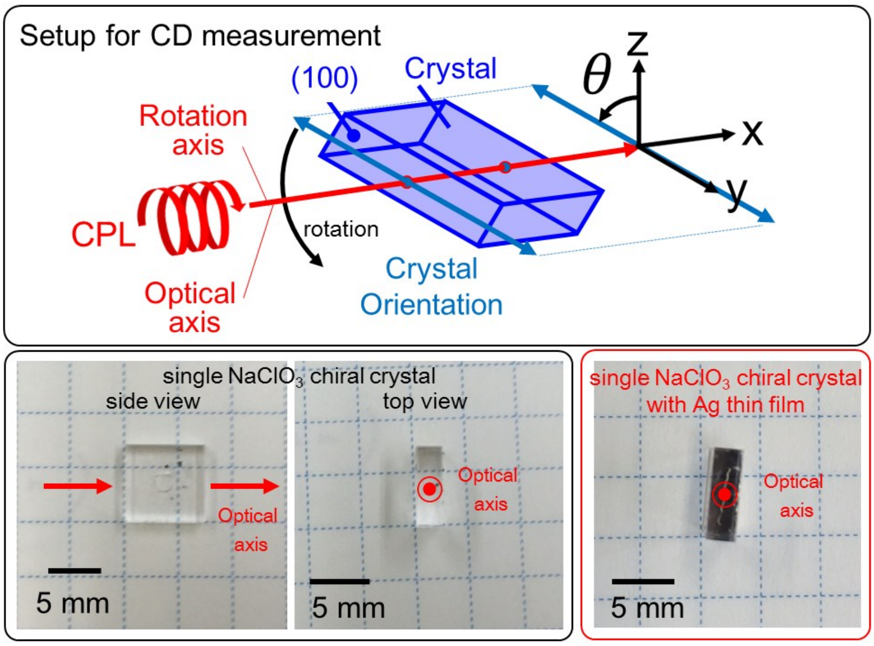

Figure ESI5. The upper schematic showing a simplified experimental configuration for CD measurement. The lower images are the samples for CD measurements. Left and middle images show a pure single $NaClO_3$ chiral crystal. Right image shows a single $NaClO_3$ chiral crystal with Ag thin film.

ESI6. The dependence of CD spectra on the crystal orientation

Figure ESI6 (a)-(h) shows the dependence of CD spectra normalized by the value of the peak at 235 nm on the crystal orientation. (a)-(f) corresponds to the case that the angles between the long side of the crystal face and the z axis are 0, 45, 90, 135, 180, 225, 270, 315^{o}, respectively. The black and red lines indicate the CD spectra of pure *d*-crystal and that of *d*-crystal with Ag thin film, respectively. The region highlighted by blue is the region where the normalized CD value of the Ag-deposited crystal positively shifts relative to that of the pure crystal and the red region is the region where the CD value of the Ag-deposited crystal negatively shifts relative to that of the pure crystal. The light red dashed line indicates the wavelength of 292.6 nm, which corresponds to the wavelength of the additional peak indicated in Figure 4 **b.** In the CD spectra of the pure crystal, the sign of the CD value from 237nm to about 300 nm changes every 90^{o} rotation, indicating the CD value originates from birefringence of crystal. Although the CD of the Ag-deposited crystal also exhibits the tendency of birefringence, shift of CD value from CD value of the pure crystal is mostly positive in the region from 237 nm to 249 nm, from 335 nm to 430 and is mostly negative in the region around 300 nm independently from the 90^{o} rotation of crystal orientation. These result shows the deposition of Ag thin film modulate CD spectra of NaClO$_3$ chiral crystal and generate additional peaks in the longer wavelength that the intrinsic CD peak at 235 nm. These additional peaks are possibly induced by the coupling between crystal chirality and the absorption of free electron in the silver thin film.

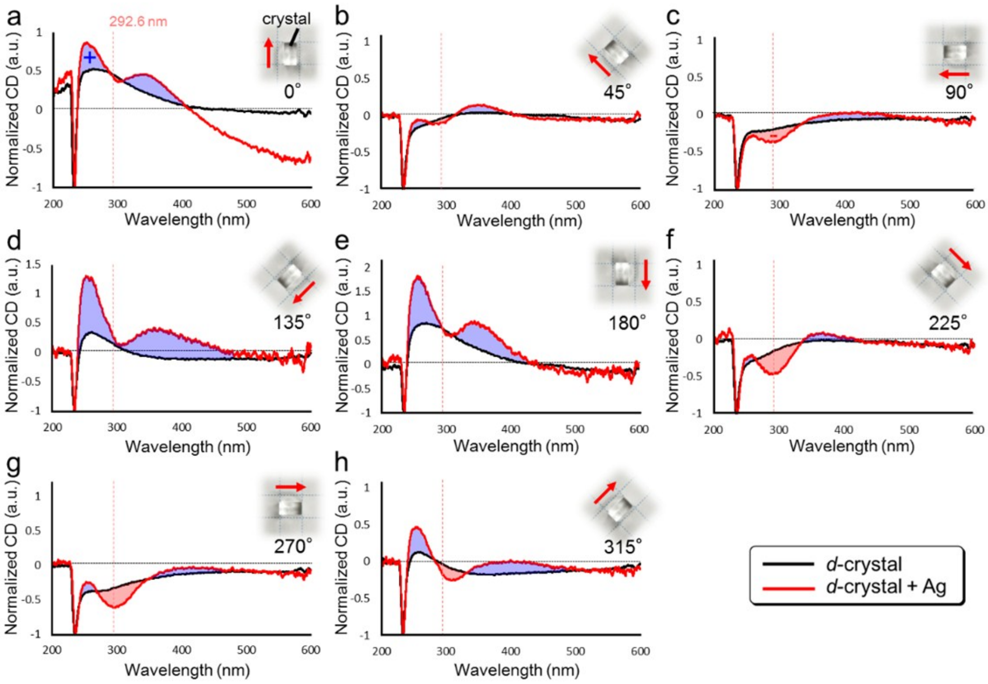

Figure ESI6. **a** to **h** shows CD spectra normalized by the CD peak at 235 nm when the rotation angles of the crystal orientation are 0, 45, 90, 135, 180, 225, 270, 315^{o}, respectively. Black and red solid lines show the CD spectra of pure d-crystal and d-crystal with Ag thin film, respectively.