Supplementary Information (SI)

Si Nanocrystal Solution with Stability for One Year

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Table S1 Liquids for PLAL of Si.

	liquid	references
hydrophilic	water	1–5
	alcohol	1,6–8
hydrophobic	alkane	7,9
	alkene	10–12
	alkyne	14,15,13
	halide	1,16
	aromatic hydrocarbon	7,9,17

Different liquids produce different passivations (i.e., water or ethanol for hydroxyl or oxide passivation, 1-octene or 1-octyne for carbon chains and partial oxides, and toluene for a graphite shell).

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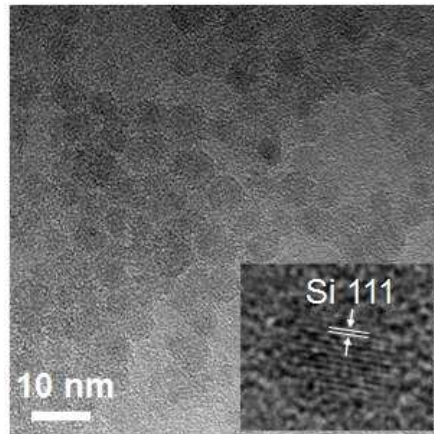(17) Intartaglia, R. et al. *Phys. Chem. Chem. Phys.* **14**, 15406–15411 (2012).

Fig. S1 High-magnification TEM image of SiNCs.

Figure S1 shows that SiNCs are approximately spherical particles. Thus, we treated the SiNC as a spherical particle for the calculation of the crystalline size from the Raman spectra.
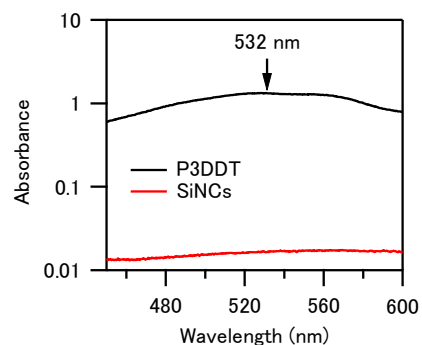
Fig. S2 Absorption spectra of P3DDT and SiNCs films. Film thicknesses of former and latter are 200 and 130 nm, respectively. The wavelength of incident light at photoconductivity measurement was set as 532 nm.

For the photoconductivity measurements of SiNCs/P3DDT hybrid films, it was ensured that the light intensity > 99% at 532 nm is absorbed by P3DDT, according to the results of absorption spectra, as shown in Fig. S2. P3DDT excitons are generated as a principle component in the hybrid film.
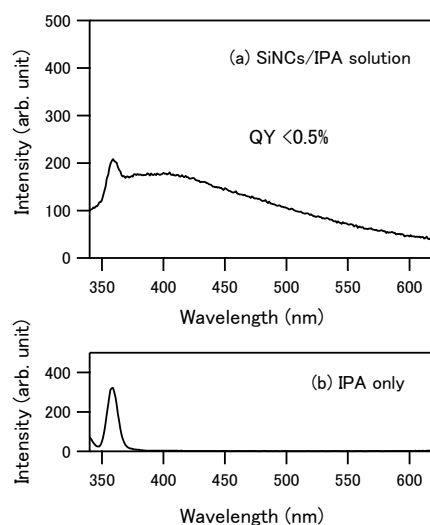
Fig. S3 Typical photoluminescence spectra of (a) SiNCs/IPA solution and (b) IPA solvent at the excitation wavelength of 325 nm. The peak at around 360 nm is attributed to the Raman band of CH stretching mode of IPA.

The photoluminescence (PL) spectrum and the PL quantum yield (QY) of the SiNCs dispersed in IPA were measured using a fluorometer (FluoroMax-4, Horiba) and a spectrometer with an integrated sphere (C11347, Quantaurus-QY, Hamamatsu Photonics), respectively. The profile of PL spectrum of SiNCs is shown in Fig. S3 (a) and its QY was estimated as $< 0.5\%$.

Note S1

The relative ratio of Si–O components $R_{\text{Si-O}}$ (%) was obtained using the following equation,

$$R_{\text{Si-O}} = \frac{n_{\text{SiO}}}{(n_{\text{SiC}} + n_{\text{SiH}} + n_{\text{SiO}})} \times 100$$

where $n_{\text{SiO}} = \sigma^{-1} \int A_{\text{SiO}} \tilde{v}^{-1} d\tilde{v}$, $n_{\text{SiH}} = \sigma^{-1} \int A_{\text{SiH}} \tilde{v}^{-1} d\tilde{v}$, $n_{\text{SiC}} = \sigma^{-1} \int A_{\text{SiC}} \tilde{v}^{-1} d\tilde{v}$,

and σ, A, and $\tilde{v}$ are the absorption cross-section, the absorbance multiplied by the wavenumber, and the wavenumber, respectively, for each vibrational band in the infrared spectrum.[1–3] Similarly, the relative ratios of Si–H and Si–CH$_x$ were obtained. The A values were obtained from the 2280-2000 cm^{-1}, 1200-800 cm^{-1}, and 1100-650 cm^{-1} regions of the SiH, SiO, and SiC components, respectively, in Fig. S4.
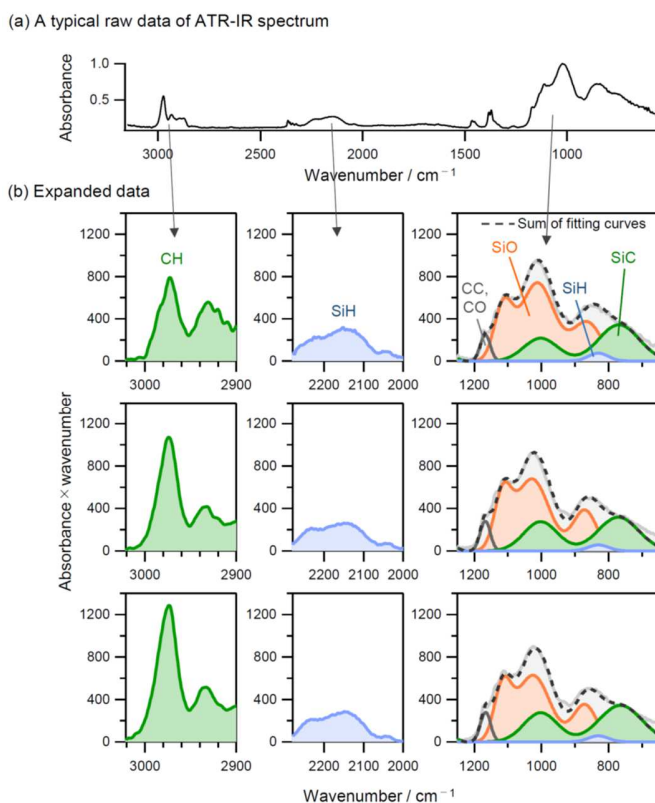

Fig. S4 FTIR spectra of the SiH, SiO, and SiC regions. For the 1200-650 cm^{-1} region, the spectra were analysed using Gaussian functions. Each bands are assigned in refs. 1-3.

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Note S2

The electronegativities of O, C, H, and Si atoms have been reported as 3.4, 2.55, 2.1, and 1.9, respectively, and were used to discuss the surface charge from passivation.[1–4] The electronegativity difference for the O atom in the Si–O bond is 1.9 – 3.4 = –1.5. Similarly, the electronegativity difference for the H atom in Si–H is –0.2 and that for the H atom in C–H is +0.45. Using the passivation ratio in Table 1, the degree of surface charge on SiNC is estimated as

degree of surface charge $= (-1.5 \times R_{Si-O}) + (-0.2 \times R_{Si-H}) + (0.45 \times R_{Si-C} \times n_{C-H})$

where R_{Si-O}, R_{Si-H}, and R_{Si-C} are the passivation ratios of each component. n_{C-H} is the number of C–H bond and was 2.5, 2.6, and 2.7 for 0.2 J cm^{-2}, 0.5 J cm^{-2}, and 0.9 J cm^{-2}, respectively. As a result, the degree of surface charge was –30, –21, and –11 for the SiNCs generated at 0.2 J cm^{-2}, 0.5 J cm^{-2}, and 0.9 J cm^{-2}, respectively, in the present study. This result indicates that the net surface charge has a negative value. This negative sign is consistent with the experimental results, i.e., the negative sign of the ζ-potential observed in Figure 2a.

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Note S3 Photoconversion efficiency to hole at the excitation wavelength of 532 nm.

The number of photons, n_{hv}, of incident light for photoconductivity measurements was 1.3×10^{12} using the equation of $n_{hv} = I/(hc/\lambda)$, where I is energy of incident light (0.5 μJ), h is Planck's constant, c is speed of light, and λ is wavelength of incident light (532 nm).

The number of holes, n_{hole}, was 6×10^9 and 3×10^{10} for P3DDT film and SiNCs/P3DDT hybrid film, respectively, using the equation of $n_{hole} = e/q$, where e is elementary charge and q is the charge of holes.

Using the n_{hv} and n_{hole}, the number of holes per the number of photons, $(n_{hole}/n_{hv}) \times 100\%$, was estimated as 0.5% and 2.3% for P3DDT film and SiNCs/P3DDT hybrid film, respectively.

Note S4 Heat dissipation time for nanoparticles

The following equation was used to estimate the heat dissipation time t of nanoparticles in a solvent,[1–3]

$$t = (R_{NP}\, \rho_{NP}\, C_{NP})^2 / (9\rho_S\, C_S\, \Lambda_S)$$

where R, ρ, C, and Λ are the radius, density, heat capacity, and thermal conductivity, respectively. The subscripts of NP and S denote the nanoparticle and solvent, respectively.

In the current study, the diameters of the final product SiNCs were 12 nm, 8 nm, and 6 nm, for low, middle, and high fluences, respectively. The heat dissipation time in IPA was calculated by substituting the nanoparticle sizes into the equation. As a result, the heat dissipation times for SiNCs of 12 nm, 8 nm, and 6 nm diameter were estimated as 4×10^{-11} s, 2×10^{-11} s, and 1×10^{-11} s, respectively.

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