Schrödinger’s red pixel by quasi-bound-states-in-the-continuum

Zhaogang Dong\textsuperscript{1,2+\dag}, Lei Jin\textsuperscript{3,4\dag}, Soroosh Daqiqeh Rezaei\textsuperscript{5}, Hao Wang\textsuperscript{5}, Yang Chen\textsuperscript{3}, Febiana Tjiptoharsono\textsuperscript{1}, Jinfa Ho\textsuperscript{1}, Sergey Gorelik\textsuperscript{6}, Ray Jia Hong Ng\textsuperscript{5\ddag}, Qifeng Ruan\textsuperscript{5}, Cheng-Wei Qiu\textsuperscript{3\ast}, Joel K. W. Yang\textsuperscript{1,5\ast}

While structural colors are ubiquitous in nature, saturated reds are mysteriously absent. This long-standing problem of achieving Schrödinger’s red demands sharp transitions from “stopband” to a high-reflectance “passband” with total suppression of higher-order resonances at blue/green wavelengths. Current approaches based on nano-antennas are insufficient to satisfy all conditions simultaneously. Here, we designed Si nanoantennas to support two partially overlapping quasi-bound-states-in-the-continuum (q-BIC) modes with a gradient descent algorithm to achieve sharp spectral edges at red wavelengths. Meanwhile, high-order modes at blue/green wavelengths are suppressed via engineering the substrate-induced diffraction channels and the absorption of amorphous Si. This design produces possibly the most saturated and brightest reds with ~80% reflectance, exceeding the red vertex in sRGB and even the cadmium red pigment. Its nature of being sensitive to polarization and illumination angle could be potentially used for information encryption, and this proposed paradigm could be generalized to other Schrödinger’s color pixels.

INTRODUCTION

Dye-free nanostructural colors offer unprecedented print resolution, fade resistance, scalable manufacturability, and viewing angle independence (1). High-resolution color printing beyond the optical diffraction limit was first demonstrated with metallic nanostructures with localized plasmon resonances, such as gold (2) and aluminum (3, 4). However, the achieved color saturation of these plasmonic nanostructures is limited partly because of ohmic losses in metals. On the other hand, dielectric nanostructures support Mie resonances (5–15) that produce color pixels with improved saturations due, in part, to abrupt spectral transitions. For instance, Si nanostructures that behave as if suspended in free space when placed on Si\textsubscript{3}N\textsubscript{4} anti-reflection layer achieve Kerker’s conditions that sharpen spectral transitions for highly saturated colors (11). Similarly, TiO\textsubscript{2} nanostructures with electric dipole and magnetic dipole resonances have demonstrated high color saturation (12) and tunability (13). Furthermore, the color saturation has been improved further by the multi-layer dielectric stacked nanostructures (14) and refractive index matching layers (16).

Despite notable progress in nanostructural colors (2–4, 11–14, 16–22), a long-standing challenge still remains in achieving highly saturated reds, where the achieved red color saturation are still within the standard RGB (sRGB) triangle in the International Commission on Illumination (CIE) chromaticity diagram, and thus, it demands a completely new design principle. Similar limitations are also observed in structural red colors in nature (23). For example, bird feathers exhibit only saturated structural blues and greens but not reds (23, 24). For instance, even the “red” feathers of parrots (Aves: Psittaciformes) have a magenta appearance, as they exhibit substantial blue and green components in the reflectance spectrum (25). In addition, the scales of longhorn beetles (26) and natural cuttlefish ink (27) have structural colors spanning the visible spectrum, with the exception of saturated red (closest being magenta). This elusive-red phenomenon could be partially explained by the underlying physics: A highly saturated red pixel requires the fundamental structural resonance beyond 600 nm in wavelength. However, resonators that support fundamental modes in red would also support higher-order modes at shorter wavelengths (i.e., 380 to 480 nm). Consequently, these high-order modes will shift the color toward blue in the chromaticity diagram, deteriorating its red saturation and eventually result in magenta instead (28). In other words, generating a highly saturated red in reflection mode requires a high reflectance of the red wavelengths (600 nm and longer) and a concomitant near-total suppression of reflectance or scattering at other wavelengths. An ideal reflectance spectrum is one with unity reflectance in the desired wavelength range and zero reflectance elsewhere, which was recently coined the Schrödinger pixel based on Erwin Schrödinger’s considerations (22, 29). Incidentally, highly saturated reds are also mysteriously rare in synthetic and natural pigments and dyes based on light absorption (23, 28), where these pigments and dyes are working under white light illumination, being different from emissive devices, such as light emitting diodes and lasers.

Here, we designed a Si nanoantenna array with two quasi-bound-states-in-the-continuum (q-BIC) modes to achieve the ideal Schrödinger’s red pixel with a high color saturation, where the high-order q-BIC modes at blue/green wavelengths are suppressed via engineering the substrate-induced leakage effect and the ohmic loss (k) of amorphous Si. In simulation, the achieved color saturation in CIE coordinate is (0.684, 0.301), which is much better than

\textsuperscript{1}Institute of Materials Research and Engineering, A*STAR (Agency for Science, Technology and Research), 2 Fusionopolis Way, #08-03 Innovis, Singapore 138634, Singapore. 
\textsuperscript{2}Department of Materials Science and Engineering, National University of Singapore, 9 Engineering Drive 1, Singapore 117575, Singapore. 
\textsuperscript{3}Department of Electrical and Computer Engineering, National University of Singapore, 4 Engineering Drive 3, Singapore 117583, Singapore. 
\textsuperscript{4}College of Electronic and Information Engineering, Hangzhou Dianzi University, Hangzhou 310018, China. 
\textsuperscript{5}Singapore University of Technology and Design, 8 Somapah Road, Singapore 487372, Singapore. 
\textsuperscript{6}Singapore Institute of Food and Biotechnology Innovation, A*STAR, 31 Biopolis Way, #01-02 Nanos, Singapore 138669, Singapore. 
\textsuperscript{\ast}Corresponding author. Email: joel_yang@sutd.edu.sg (J.K.W.Y.; chengwei.qiu@nus.edu.sg (C.-W.Q.); dongz@imre.a-star.edu.sg (Z.D.)
\textsuperscript{\dag}These authors contributed equally to this work.
\textsuperscript{†}Present address: Institute of High Performance Computing, A*STAR, 1 Fusionopolis Way, #16-16 Connexis, Singapore 138632, Singapore.
the red in the sRGB triangle or the well-known “cadmium red” pigment with the respective CIE coordinates of (0.64, 0.33) and (0.621, 0.32) (30). The geometry optimization of q-BIC antenna is based on a gradient descent approach integrated with finite-difference time-domain (FDTD) simulation to search for the brightest and most saturated red via suppressing reflectance at blue and green wavelengths. Distinct from the more common x-polarized q-BIC modes in the literature (31–33), here, we introduce y-polarized q-BIC modes to achieve a better red color saturation and higher reflectance. Experimentally, the achieved red saturation indeed exceeds the sRGB triangle with a CIE coordinate of (0.654, 0.301) while maintaining a high reflectance of ~80%. In addition, the achieved color saturation has a strong dependence on the incidence angle and polarization due to the nature of q-BIC resonances. The achieved highly saturated red could be potentially scaled up, e.g., via deep ultraviolet lithography and nanoimprint lithography (34), to reach the dimensions of reflective displays based on multilayer film configuration (35) and lead to potential applications of compact red filters (36), highly saturated reflective displays (22, 37), nonlocal metasurfaces (38), and miniaturized spectrometers (39).

RESULTS
Design of BIC antenna array for achieving the ideal Schrödinger’s red pixel
Figure 1A presents the reflectance spectrum of an ideal Schrödinger’s red pixel, which has a flat top profile with a sharp transition from “stopband” to “passband.” In comparison, Fig. 1A also presents the schematic illustration of a typical nanophotonic optical resonance, which cannot achieve highly saturated red because of the following two reasons. First, when the nanophotonic optical resonance has a fundamental mode at the red wavelength, it usually has the higher...
order mode at the blue wavelength (11, 12). Second, the broad resonances are substantially different from the Schrödinger’s type resonance with steep edges.

On the other hand, BIC is an emerging concept in nanophotonics (31–33, 40–47), and this BIC mode could potentially provide highly saturated reds. As the true BIC mode is optically inaccessible (40), here, we use the q-BIC mode with broken symmetry (31, 32, 40). Figure 1B presents the simulated reflectance spectrum of the designed q-BIC antenna array with two BIC resonances at y-polarized incidence condition, where these y-polarized q-BIC resonances were not explored so far. Here, the merging of two q-BIC resonances will give rise to steep edges to enable sharp transition from stopband to passband. Meanwhile, the merging of two q-BIC resonances will generate considerable full width at half maximum, which is required for saturated red with high brightness. Although one-dimensional (1D) photonic crystal is able to achieve a bandstop with a sharp transition too (48), the blue color wavelength component was not fully suppressed and it has a limited printing resolution due to the multilayer design with the infinite extension along x and y directions.

The simulated reflectance spectrum in Fig. 1B was obtained on the basis of a careful optimization of the q-BIC antenna design with the corresponding 3D schematic as shown in Fig. 1C. This q-BIC antenna is based on amorphous Si, with n and k values as shown in fig. S1. The designed unit cell consists of a pair of elliptical amorphous Si nanostructures with a long-axis L and a short-axis W. Its long axis has an angle θ with respect to the x axis. The pitch along the x and y direction are denoted as λx and λy, respectively. These parameters will be used in the optimization algorithm as discussed later. In addition, Fig. 1D presents the mode patterns of two q-BIC modes being excited on the antenna array. Moreover, Fig. 1E presents the achieved color saturation in a small section of the CIE 1931 chromaticity diagram, with the CIE coordinates of (x, y) = (0.684, 0.301) in simulation and (0.654, 0.301) in experiment, where the achieved red saturation is beyond the sRGB triangle. These achieved reds are more saturated than the cadmium red pigment, known for producing vibrant reds in modern art, with the CIE coordinate of (0.684, 0.301). The optimized parameters are λx = 47.9°, λy = 436 nm, λy = 432 nm, and HSi = 360 nm.

Next, we present the detailed gradient descent algorithm as integrated with FDTD simulation package to design and optimize the geometric parameters of the q-BIC antenna array. In Fig. 2A, our first cost function is the distance “D” on the CIE diagram from the achieved structural color coordinate to the red apex of the CIE plot, which is defined here as the reddest possible red corresponding to a monochromatic light source with a wavelength of 700 nm. Unfortunately, a surface with a narrow but perfect reflectance peak at 700 nm would result in a nearly black reflective appearance. A smaller D value corresponds to a more saturated red. In addition, we introduced a second cost function to mitigate the tendency of the gradient descent algorithm to be stuck at a local minimum. The second cost function maximizes the “CIE x coordinate” so that the optimization algorithm will find the optimized geometry toward achieving the CIE plot rim at the red corner. The cost functions will be toggled from one to the other when the optimization of one cost function is stuck at a local minimum.

The detailed algorithm flow is shown in Fig. 2B. Here, the cross-sectional profile of the optimized Si antenna is based on the experimental result. For instance, a typical cross-sectional profile of the fabricated Si nanostructures is shown in fig. S2, where the etched Si nanostructures do not have perfectly vertical cross-sectional profiles because of dry etching conditions. Instead, we observe two segments. The top ~154-nm segment [i.e., the part close to the hydrogen silsesquioxane (HSQ) resist] has as ~90° side wall profile, while the bottom segment tapers outward to a trapezoidal shape with side wall angle of ~82°. The modeled antenna cross section is shown in fig. S3.

The parameters of the antenna design are represented as a 1D vector P = [L, W, θ, λx, λy, HSi], where these geometric parameters are shown in Fig. 1C. In addition, λx and λy denote the pitch along x and y directions, respectively. We manually generated the initial set of parameters to bring it within the red region of the CIE map.
On the basis of this initial starting input parameters, FDTD simulations were then carried out to simulate the optical field distribution and reflectance spectrum, which was used to calculate the coordinate \((x, y)\) in the chromaticity diagram.

In addition, the cost function \(F\) will be chosen between the first cost function \(D\) and the second cost function “x.” In the first step, we set “\(F = D^x\)” to minimize \(D\) for highly saturated red. After that, its first-order derivative \(\frac{\partial F}{\partial x}\) was numerically calculated for each variable \(P_i\) based on \(\frac{F(P_i + \Delta P_i) - F(P_i)}{\Delta P_i}\). Last, we obtain the optimized parameters for the minimum value of \(F\) based on the gradient descent approach after a number of iterations, i.e., 60 steps, to get a convergent result. Once the optimization is stuck at a local minimum, the cost function \(F\) will be switched to “\(F = x\)” in the next round of optimization for another 20 iterations, until reaching another local minimum. The switching process of the cost function continues until the optimization result does not change anymore. A more detailed explanation of this algorithm flow is shown in fig. S4, where this gradient descent approach could be potentially improved by incorporating the topology optimization in nanophotonics (49). On the basis of our experience, HSQ resist remains on the silicon nanostructures after fabrication with a thickness of ~60 nm, which was kept constant in calculations throughout the manuscript. This 60-nm-thick HSQ enhances the color saturation slightly by suppressing reflectance in the blue wavelength, as seen in simulation results in fig. S5.

Next, we performed the search for the optimized geometry under both \(x\)- and \(y\)-polarized incident conditions. First, we start our analysis with the q-BIC resonance excited along \(y\) polarization. To our knowledge, these \(y\)-polarized q-BIC resonances have not been studied so far for the nanoeclipse array. In the optimization process, the initial starting point of the geometrical parameters are \(L = 270\) nm, \(W = 100\) nm, \(\theta = 65°\), \(\Lambda_x = 460\) nm, \(\Lambda_y = 420\) nm, and \(H_{Si} = 350\) nm. Figure 2C presents the corresponding optimization trajectory in the CIE chromaticity diagram after 120 iterations, where the corresponding switching between \(F = D\) and \(F = x\) is shown in fig. S6. The optimized parameters of Si nanostructures achieving the highly saturated red are \(L = 235\) nm, \(W = 85\) nm, \(\theta = 47.9°\), \(\Lambda_x = 436\) nm, \(\Lambda_y = 432\) nm, and \(H_{Si} = 360\) nm. The corresponding CIE 1931 chromaticity diagram is shown in Fig. 2C with a CIE coordinate of \((0.684, 0.301)\), where the reflectance spectrum is shown in Fig. 1B with two prominent q-BIC peaks at red wavelengths. Here, these two red q-BICs do not have the high-order diffraction since these red wavelengths are above the wavelength criterion \(\lambda_0\), which can be calculated by

\[
\lambda_0 = n_{\text{substrate}} \times \max \{\Lambda_x, \Lambda_y\}
\]

In addition, fig. S7 presents the transmittance and absorbance spectra, where figs. S8 and S9 present the evolution of reflectance spectra with respect to different tilt angle \(\theta\) of the nanoeellipses and the influence of the side-wall slant angle. It shows that the reflection peaks due to the q-BIC resonance will slowly disappear when \(\theta\) is changed from 47.9° to 0°. At the same time, the corresponding Q factor will be increased as shown in fig. S10. Similarly, the optimization for \(x\)-polarized incidence condition is shown in the figs. S11 and S12 to achieve a saturated red with a CIE coordinate of \((0.678, 0.307)\) but with a lower reflectance as compared to the \(y\)-polarized design.

**Experimental fabrication and characterization**

Experimentally, we choose the design as optimized under \(y\)-polarized condition because of its higher reflectance and better color saturation as compared to the \(x\)-polarized case. We fabricated the Si nanostructures by using high-resolution electron-beam lithography (EBL) and followed by reactive-ion etching (see details in Materials and Methods). Briefly, amorphous Si layer was grown onto the quartz substrate by using plasma-enhanced chemical vapor deposition (PECVD) method. After that, 100-nm-thick HSQ resist (Dow Corning, XR-1541-006; 6% dissolved in methyl isobutyl ketone) was spin-coated onto the sample surface at 5000 rpm, followed by EBL and dry etching processes (50). The detailed fabrication process is shown in fig. S13 with the corresponding process parameters described in Materials and Methods. Figure 3 presents the corresponding nanofabrication and characterization results.

To minimize the undesired reflectance from the bottom quartz-air interface, we coat it with an absorbing pigment analogous to *Morpho* wing scales that incorporate melanin below the multilayered ridges (51, 52) to absorb stray light, which would otherwise scatter back and substantially reduce color saturation (52). In our experiment, black ink was coated onto the back side of the quartz substrate, as shown in the corresponding schematic in Fig. 3A. The deposition of black dye is carried out by scribing “Sharpie Permanent Marker (model number: MK-SP-FINE-BLK)” onto the back side of quartz substrate uniformly. In addition, Fig. 3 (B and C) presents the side view and top view of the scanning electron micrograph (SEM) images of the fabricated Si nanoeclipse antenna array.

Figure 3D presents the measured reflectance spectrum, and the inset figure is its optical micrograph of the final sample (see Materials and Methods). This reflectance spectrum was measured by a microspectrophotometer [CRAIC UV-VIS-NIR QDI 2010; ×5 objective lens; 0.12 numerical aperture (NA)], and the corresponding CIE coordinate was then calculated on the basis of the reflectance spectrum (11). This red saturation is beyond the sRGB triangle in the CIE chromaticity diagram as shown in Fig. 3E, with the corresponding CIE coordinate of \((0.654, 0.301)\). In comparison, fig. S14 presents the measured reflectance spectrum for the same red pixel without the black dye at the back side of the quartz substrate, where the corresponding CIE coordinate is only \((0.613, 0.301)\). It shows that this additional black dye on the back side of the quartz substrate is able to improve the red saturation because it is able to reduce the overall background signal in blue and green color regimes of the reflectance spectrum. The corresponding hue values for the experimentally measured and simulated reflectance spectra are 356.8° and 356.9°, respectively.

We also investigate the influence of incidence polarization, NA, and viewing angle on the achieved color saturation. Figure 4A presents the measured reflectance spectra for the same red pixel as fabricated in Fig. 3 by using the ×5 objective lens (0.12 NA) under \(y\)-polarized incidence condition. The corresponding CIE coordinate shifts to \((0.593, 0.316)\) as shown in Fig. 4B, where the maximum reflectance is reduced to ~36%. This measurement demonstrates that our red pixel based on q-BIC resonance is strongly dependent on the incident light polarization. In addition, Fig. 4 (C and D) presents the investigation on the influence of NA on the achieved color saturation under \(y\)-polarized incidence condition. It shows a reduced color saturation with increasing NA, with a shift in corresponding CIE coordinates in Fig. 4D. Thus, the most saturated reds are seen within a narrow collection angle and illumination angle.
(i.e., a solid angle $\Omega$ of $6.9^\circ$), corresponding to an NA of 0.12. The dependences of polarization and incident angles are also evident from the angle-resolved reflectance measurements at back focal plane as shown in fig. S15. Furthermore, because of the antenna array effect, the designed BIC antenna with the ultrahigh red saturation here is iridescent. Its color will be changed slightly when the view angle is changed, where the corresponding detailed comparisons are shown in fig. S16 with the tilt angle of 0° and ±15°. Therefore, these characterization results in Figs. 3 and 4 show that ultrahighly saturated reds are observed under specific conditions, i.e., $y$-polarized illumination, the narrow angle of incidence, and zero tilt angle.

In our design, we manage to suppress the high-order q-BIC modes at blue and green wavelengths by exploring the following two effects, i.e., intrinsic absorption coefficient $k$ of amorphous silicon and the substrate-induced leakage effect, where the detailed simulation comparisons are shown in fig. S17. For instance, fig. S17B shows that high-order q-BIC modes will appear if the intrinsic absorption coefficient $k$ of amorphous silicon is set to zero since the q-BIC resonances are very sensitive to $k$. Moreover, fig. S17 (C and D) presents the investigation when the quartz substrate is absent, where many high-order q-BIC modes are emergent. It is because that the presence of a high-index substrate will break the out-of-plane symmetry of the optical system and open up high-order diffraction channels for these q-BIC modes (53). In this way, the at-Γ BICs protected by the in-plane symmetry will be destroyed and converted to leaky resonances.

We applied the following protocol to characterize and calculate the achieved CIE 1931 coordinates based on the measured reflectance or transmittance spectra. The approach could be useful in...
standardizing future work for objective benchmarking. First, the
calculation of CIE 1931 coordinate requires the full spectral informa-
tion from 380 to 780 nm (anything less would artificially boost
saturation). To simplify matters and avoid the influence of the light
source, the sample is assumed to be illuminated by a perfect broad-
band light source with unity relative power from 380 to 780 nm.
Then, on the basis of this full spectral information, the CIE coordi-
nates of the color pixels could be accurately calculated on the basis
of the eqs. S1 to S5 in fig. S18.

**DISCUSSION**

In addition to red, the presented q-BIC antenna could be extended
to other colors, as shown in fig. S19 for the case of green. On the
other hand, it is challenging to use q-BIC resonance to achieve the
highly saturated blue pixel based on amorphous silicon. The reason
is that amorphous silicon has a relatively large ohmic loss (i.e., k)
at the blue wavelength of below 480 nm (see fig. S2). As a result, the
absorption-induced optical loss dominates radiative losses. Thus,
engineering q-BIC resonance to achieve highly saturated blue pixel
may require other dielectric materials with lower ohmic loss at blue
color wavelength, such as TiO$_2$ (12). Moreover, the pixel size based
on q-BIC resonance cannot be reduced as much as the typical struc-
tured color elements based on the local resonances (11) because the
q-BIC resonance is an array effect, which requires a minimum
number of unit cells of ~11 × 11 elements in the case of Si (40).

Here, we present a Si antenna design with q-BIC resonances to
achieve highly saturated red, which is beyond the sRGB triangle in
the chromaticity diagram. The algorithm used aided the discovery
of new y-polarized q-BIC modes. These fundamental and higher-
order q-BIC modes closely placed within the spectral passband of
the ideal Schrödinger red provide the desirable sharp transition
necessary for Schrödinger color saturation. To the best of our
knowledge, the results represent the highest red saturation achieved
by nanostructures with full spectral characterization across from
380 to 780 nm. Notably, because of the angle- and polarization-depen-
dent nature of the color, the presented design could be exploited
for unique optical effects that are absent in pigments for
polarization encrypted anti-counterfeiting applications (34). More-
over, different configurations based on single asymmetric dielectric
nanostructures could be explored further with smaller footprints
toward higher printing resolution (31, 54).

**MATERIALS AND METHODS**

**Numerical simulations**

A commercial software (Lumerical FDTD Solutions) was used to do
the FDTD simulations. Along the x/y directions, periodic boundary
conditions were used, and perfect matched layer was used along the
z direction. The $n$ and $k$ values of the amorphous Si were measured
by ellipsometer, as shown in fig. S1. On the basis of the Lumerical
script and achieved reflectance spectra by simulation, the CIE 1931
chromaticity diagram is calculated by the color matching functions,
where the detailed formulation could be found in fig. S15.

**CVD growth of Si film, nanofabrication, and Si etching**

PECVD method was used to grow the amorphous Si film onto
quartz substrate. The etching masks were fabricated on the basis of
100-nm-thick HSQ, which was obtained by spin coating the resist
(Dow Corning XR-1541-006) at 5000 rpm. The layout file of nano-
ellipse array was created by using the software AutoCAD, and then,
this file was converted into the exposure file by another software
Beamer (GenISys GmbH). The EBL (Elionix) was then conducted
with an acceleration voltage of 100 keV, beam current of 200 pA,
and an exposure dose of ~12 mC/cm$^2$. The sample was then developed
by a NaOH/NaCl salty solution (1% weight/4% weight in deionized
water) for 60 s and immersed in deionized water (55). Next, the
sample was immediately rinsed by acetone and isopropanol alcohol
and dried by a continuous flow of nitrogen. Si etching was then
carried out by using inductively coupled plasma (Oxford Instruments
Plasma Lab System 100), with Cl$_2$ gas chemistry at 40°C.

**Characterizations**

The optical reflectance spectrum was measured by using a CRAIC
microspectrophotometer QDI 2010 (×5 objective lens, Zeiss A-plan
with an NA of 0.12) under the illumination of polarized broadband
light source (75-W xenon lamp). The absolute reflectance spectra
were obtained, where we calibrated our reflectance measurements
on a certified calibration aluminum sample from CRAIC Technol-
ogies. Moreover, for the acquisition of the optical microscope image
in Fig. 3, a white color balancing was first carried out on the calibra-
tion aluminum sample. The images were then taken by using a
microscope (Olympus MX61) with the “analySiS” software, a ×5 objec-
tive lens (MPlanFL N; 0.15 NA), a camera (Olympus SC30) with an
integration time of 20 ms, and a halogen light source (ULH100 3)
with a linear polarizer. The SEM images were taken with an electron
acceleration voltage of 10 keV (SEM, Elionix).

**SUPPLEMENTARY MATERIALS**

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