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Published in: Optical Materials Express

DOI: 10.1364/OME.9.000717

Publication date: 2019

Document version: Final published version

Citation for published version (APA):
Deshpande, R. A., Roberts, A. S., & Bozhevolnyi, S. I. (2019). Plasmonic color printing based on third-order gap surface plasmons. Optical Materials Express, 9(2), 717-730. https://doi.org/10.1364/OME.9.000717

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Download date: 15. May. 2021
Plasmonic color printing based on third-order gap surface plasmons [Invited]

Rucha A. Deshpande,1,* Alexander S. Roberts,1 and Sergey I. Bozhevolnyi1,2

1 Centre for Nano Optics, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark
2 Danish Institute for Advanced Study, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark
*rad@mci.sdu.dk

Abstract: Metasurfaces for color printing rely on the resonant interaction of light with nanostructures, which translates to small physical resonator sizes for visible wavelengths, thus demanding very high fabrication accuracy that necessitates low-throughput processes and prevents the economical large-scale coloration and printing. Through the use of third-order gap plasmon resonances, element sizes necessary for visible resonances can be scaled to be compatible with several large-scale techniques for nanofabrication while retaining the capability of generating vivid colors. We demonstrate this using a 450-nm-periodic gold nanodisc array on SiO2 of different thicknesses (20, 40 and 50 nm) atop a thick gold substrate - a composite structure supporting gap plasmon resonances for both linear polarizations. A third-order resonance occurs for discs of 300 nm diameter and larger, while first-order resonances occur below 100 nm for red light (630 nm). By fabricating nanodiscs of different diameters, we observe vivid shades of red and green colors, with dark shades for third-order gap plasmon nanodiscs resonators due to inherent losses and bright shades for small discs of fundamental resonance counterparts. The third-order gap plasmon based nanodiscs are further spectrally characterized and tested for uniformity and reproducibility by demonstrating printed patterns. The significant improvement in both size and color range using the approach of higher-order based resonances can have a significant impact on the practical applicability and economic feasibility of plasmonic color printing.

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1. Introduction

Metasurfaces are increasingly popular for color printing due to their ability to reduce the pixel size to the subwavelength regime using ultra-thin structured films, allowing for high resolution and low bulk volumes, which cannot be achieved using conventional dyes [1]. Since, metasurface elements are composed of nanostructures that resonantly interact with light to manipulate phase, amplitude, and polarization in the far-field, their geometrical parameters depend on the operation wavelength. For plasmonic structures that rely on strong electromagnetic field interactions with metal-dielectric-metal configuration that support gap surface plasmons (GSPs), efficient and versatile applications ranging from the visible to the near-infrared range including polarization splitting and polarimetry [2–10], waveplates [11–15], multifunctional focusing [16–18], integrated functionalities of splitting and surface waves [19,20], holograms [21–23] and color printing [1,24–27] have been studied. Among them, applications such as holograms and color printing, which can be perceived by human eyes are unavoidable in the visible wavelength range. Thereby, color printing applications have been extensively studied to have best possible features of color vibrancy, viewing angle independence and stability using both plasmonic and dielectric metasurfaces. Plasmonic color printing applications for different types of resonators such as nanodiscs and holes composed of various materials including gold, silver and aluminium have been studied to achieve wide color range [28–30]. Considering the metal-insulator-metal
configuration, another type of geometry which comprises of varying thickness of dielectric layer to tune the far-field spectra based on interference principle for both reflection [31] and transmission [32] has been studied. However, the continuous thin dielectric layer for GSP based configuration used in our study reduces complexity and allows for spectral colors to be tuned using single layer of nanodisc arrays fabricated by one-step-lithography process. New studies considering all dielectric resonances based metasurfaces composed of materials like silicon discs on a index-matched substrate and titanium dioxide discs continue to enhance the color gamut as well as viewing angle independence, which is difficult to achieve due to inherent damping in plasmonic resonances [33–35].

However, visible wavelength metasurfaces are restricted to low throughput applications since the meta-element geometry scales to small dimensions less than 100 nm. High-resolution and costly fabrication techniques commonly used for such sizes are electron beam lithography (EBL) and focused ion beam milling (FIB). Moreover, complex fabrication facilities like atomic layer deposition or wafer bonding are required for dielectric materials. To fulfill high-throughput demands for color printing, methods like imprint lithography, roll-to-roll printing and alternative methods of laser writing and laser post-processing have been studied [26,36–39]. Specifically, fabrication-wise less complicated structures like polycrystalline silicon for holograms have been studied for manufacture of small size meta-elements, however, these studies are restricted to few types of metasurfaces [40]. Recently, a more general approach to scale the meta-elements to relatively large sizes, though still smaller than operating wavelength using higher-order based GSP resonances (GSPRs) has been studied. Due to relative losses, it is unsuitable for efficiency-demanding applications like polarization splitting [41]. However, for plasmonic color printing, the relative increase in size and its associated losses can achieve a notably wide range of colors with the advantage of reduced fabrication constraints.

In our study, we explore the approach of higher-order GSPRs, which are supported by metal-dielectric-metal interfaces, in the form of standing waves for color printing applications. We use gold metal nanodiscs on top of a thin dielectric layer of silicon dioxide (SiO$_2$) atop a thick gold layer, a structure which supports GSPRs in the visible for a 450-nm-period for both linear polarizations. Optical measurements of uniform nanodisc arrays show that reproducible vivid shades of reds and greens are obtainable from compact, large period nanodiscs with third-order GSPRs for large nanodiscs (diameter 325 nm) and the first-order counterpart for small nanodiscs <100 nm. To test the quality and reproducibility of the nanodiscs for practical color printing applications, we replicate our University logo (SDU) using the nanodiscs arrays described above. The general approach of higher-order resonances can be further studied for different types of materials, geometries, and resonances, to enhance the features of both color vibrancy and ease-of-manufacture, which can have a significant impact on large-scale structural color printing.

2. Design and simulations

We begin with a metal-dielectric-metal GSP configuration, illuminated with normally incident light as shown in Section 2(a), a configuration which has been studied extensively in the past. The nanodisc geometry allows for polarization-independent phase and amplitude modulation of reflected light [42]. The top layer is composed of 50 nm thick gold nanodiscs above a thin dielectric layer of SiO$_2$ followed by an optically thick gold layer. The thickness of SiO$_2$ layer is varied (20, 40 and 50 nm) to study its influence on the color saturation obtained by nanodiscs of different diameters.

The configuration supports surface plasmons, as the incoming electromagnetic field excites the GSP mode at the metal-dielectric-metal interface, which is reflected at the terminations of the nanodiscs, thereby producing standing wave resonances. Thus, they can be appropriately scaled to obtain large disc dimensions, which can be explained by the Fabry-Pérot (FP) resonance
where, $D$ describes the diameter of the nanodisc, $k_0$ is the vacuum wave number, $n_{gsp}$ is the real part of the effective refractive index of the GSP, which depends on the material parameters including thicknesses, $p$ is the integer defining the order of the GSP mode, $\phi$ is an additional phase accumulation that accounts for the fact that GSPs are not reflected from the exact physical boundaries of the metal-dielectric-metal configuration since part of the field extends outside the structure.

A period ($\Lambda$) of 450 nm is chosen to support both a third-order GSPR ($p = 3$) for larger nanodiscs and its fundamental counterpart ($p = 1$) for smaller nanodiscs at visible wavelengths, shown schematically in Fig. 1(a). This is confirmed by mapping the electric and magnetic fields of modes positioned spectrally at 630 nm for a large nanodisc diameter of 325 nm and a small nanodisc diameter of 75 nm as shown in Figs. 1(c) and 1(d) for a SiO$_2$ thickness 40 nm. The corresponding GSP modes for different SiO$_2$ thicknesses of 20 and 50 nm are also studied and shown in Appendix, Figs. 5(a) and 5(b), respectively. By comparing the GSP modes for different thicknesses, it is clearly observed that for the increase in the SiO$_2$ layer thickness, the overall reflectivity decreases leading to darker color appearance. We use the commercially available finite-element solver COMSOL Multiphysics to simulate the mode structure using the permittivity of gold obtained by Johnson and Christy [44] and the refractive index of SiO$_2$ taken to be 1.45. By varying the diameter of nanodiscs, we engineer the amplitude and phase of GSPR to achieve color saturation for desired visible wavelengths in the far-field, which is schematically shown in Fig. 1(b).

3. Results

In this section, the characterization of nanodiscs is performed and the colors obtained are implemented in printing a design pattern. This is described through appropriate subsections where, the optical, spectral features of different diameter nanodiscs and color printing of design pattern are individually explained.

3.1. Performance of nanodiscs

The first experimental verification is performed by fabricating uniform nanodisc arrays of different diameters ranging from 50 nm to 425 nm with 25 nm increments and a period of 450 nm in arrays of 20 $\mu$m length sufficient for imaging. The nanodisc arrays are fabricated on 20, 40 and 50-nm-thick SiO$_2$ dielectric layers. We obtain CCD images of nanodiscs observed by bright-field optical microscope using a 50$\times$ objective with the aperture stop at a minimum diameter to ensure close to normal illumination. The images are shown in the form of the color palette in Fig. 2(a). The fabrication quality is shown by SEM images in Figs. 2(b), 2(c) and 2(d) for nanodisc arrays of diameter 100, 250 and 400 nm, respectively, on different SiO$_2$ thicknesses marked by dashed squares.

From Fig. 2(a), it emerges that bright colors with distinct color variation are observed for increasing thickness of SiO$_2$ layer. It is clearly observed that the small diameter nanodiscs supporting fundamental GSPRs are fabricated with sufficient accuracy, as they show homogeneous light colors in the far-field. The colors become relatively dark and increasingly distinct for increasing diameter of nanodiscs corresponding to the third-order GSPR, which indicates an enhancement in the degree of color saturation. The distinct colors appear to be dark, which can be due to Ohmic losses associated with gold in the visible along with the higher-order losses, which can be further improved by using suitable plasmonic materials and geometries in the visible.

The resulting color palette, comprising light and dark shades of primarily red, yellow and green colors, is obtained for nanodisc diameters in a single unit cell (450 nm) with appropriately
Fig. 1. First and third-order nanodiscs GSPRs for color printing. (a) Schematic of a gold nanodisc (diameter D) atop a thin (thickness $t_s$) SiO$_2$ layer and bottom optically thick gold layer in period ($\Lambda$) of 450 nm, illuminated with normal incidence. (b) Schematic of nanodisc arrays of different diameters reflecting colored light in the far-field. (c) Electric and magnetic fields of a GSPR supporting a first-order resonance ($D = 75$ nm) and (d) third-order resonance ($D = 325$ nm).

The nanodisc spectra are measured in the wavelength range of 400-780 nm by directing the light reflected from the nanodisc arrays to a visible spectrometer described in detail in the Methods.
The reflectance of nanodiscs atop a 40 nm SiO$_2$ layer for disc diameters of 75 to 125 nm and 300 to 350 nm, corresponding to first and third-order resonances, respectively, are experimentally measured (Fig. 3(a)). The correspondingly simulated reflectance spectra using finite-element simulations for normally incident light are shown in Fig. 3(b). A conversion of the spectra to CIE xy coordinates is shown in Figs. 3(c) and 3(d), respectively. The color palette of the measured nanodiscs as seen in bright-field microscopy is shown above the figures, where the GSPRs investigated in Figs. 3(a) and 3(b) are outlined corresponding to their respective graph in the figures. The reflectance spectra experimentally measured for all nanodisc diameters are shown in Appendix, Figs. 6(a) and 6(b) and the corresponding simulated spectra are shown in Appendix, Figs. 7(a) and 7(b). In addition, we study the reflectance spectra of different SiO$_2$ thicknesses for a selection of nanodisc diameters (D = 75, 100, 325 nm) by simulations to directly inspect the influence of the dielectric layer thickness on discs of constant diameter. The reflectance plots of 20 and 50 nm SiO$_2$ thicknesses are shown in Appendix, Figs. 8(a) and 8(b), respectively. Note that although we use normally incident light, we do not see significant influence on the far-field spectra due to the larger period chosen to accommodate nanodiscs of third-order GSPRs. We confirm this by collecting the spectra of a selected nanodisc array through objective (NA 0.95 (50×)), which collects the diffracted light from smallest wavelength used in our experiments and another objective (NA 0.5 (20×)) for smaller angles. We find no significant difference in the reflectance measured for spectra collected from both objectives, which is shown in Appendix, Fig. 9.

From the bright-field reflectance spectra shown in Fig. 3(a), several main features of the GSPRs emerge. Above 500 nm, the reflectance rises for all nanodiscs, and it is in this range that the GSPRs can have a significant influence on the optical characteristics of the metasurface. The nanodisc diameters giving rise to first-order resonances in the visible show reflectance dips with a
Fig. 3. Optical characterization of nanodiscs. (a) Experimentally measured reflection spectra of nanodisc arrays on 40 nm SiO$_2$, collected with a ×50 objective (NA = 0.8) and normalized against the reflection from a silver mirror. Solid curves correspond to first-order GSPR with small diameter nanodiscs and dash curves for third-order GSPR based large nanodiscs. (b) Simulated spectra of first-order and third-order GSPR nanodiscs. (c) CIE xy color plots based on experimentally obtained and (d) simulated spectra of all nanodiscs. The circles represent the diameters ranging from 50 nm to 425 nm with increasing disc diameters represented by circles of increasing brightness.

FWHM of approximately 50 nm while the GSPRs with visible third-order resonances have wider dips in reflectance of roughly 80 nm, due to their increased round-trips losses and associated damping of the GSP mode. For an increase in the nanodisc sizes, the absorption wavelength increases almost linearly retaining the same FWHM. This feature is observed individually for nanodisc diameters in the 75-125 nm range and 300-350 nm range, which corresponds to first-order ($p = 1$) and third-order ($p = 3$) GSPRs, respectively. A striking difference is observed, where the reflection is not completely suppressed for first-order GSPR nanodiscs, which show bright colors in the far-field as seen from outlined color blocks in the palette. On the contrary, the third-order GSPRs exhibit a stronger absorption due to a better match between their absorption cross-sections and the chosen unit cell size, which gives rise to distinct colors as seen from the
color blocks outlined with dashes. This is in contrast to the first-order GSPR nanodiscs that have an absorption cross-section that is significantly smaller than the unit cell size and, therefore, do not lead to full absorption. Moreover, for the first-order GSPRs, the absorption wavelengths vary to a higher degree for small (absolute) changes in the diameter, compared to third-order GSPR nanodiscs. Thus, nanodisc arrays manufactured to significantly higher precision are needed to obtain the desired spectral features, and corresponding color variation, using first-order visible GSPRs. Similar features are also observed for different SiO$_2$ thicknesses with the increase in overall reflectivity for small thickness. However, the absorption cross-section for 20 nm SiO$_2$ thickness is weak even for third-order GSPR, which shows why distinct colors are not observed in the far-field. In comparison, the absorption cross-section is stronger for 50 nm SiO$_2$ thickness, but low overall reflectivity results in the far-field colors appearing dark. This can be seen from the simulated spectra shown in elementary Figs. 8(a) and 8(b).

Away from both resonances, the reflectance spectra of nanodiscs remain relatively constant, with a nonlinear increase in absorption wavelengths for increasing diameters. This includes the diameters from 150-225 nm and 375-400 nm, which are shown in Appendix, Fig. 6. In all the spectra, a strong absorption band at wavelengths below 500 nm, is caused by inter-band transitions of gold leading to high Ohmic losses in the material. The low reflectance also explains why it is not possible to observe blue and violet colors when using gold-based GSPRs.

The experimentally measured spectra as explained above agree qualitatively with simulated spectra as shown in Fig. 1(b) and Appendix, Fig. 7. The simulations are performed considering ideal conditions of plane wave illumination on nanodiscs without the practical imperfections like surface roughness, and adhesive titanium layers. To account for the grain boundary losses for nanodiscs, we consider the optical properties of gold with three times increased imaginary part of gold. See Methods section for more details. The simulated spectra show better correspondence overall. However, for the first order GSP resonance corresponding to small (< 100 nm) nanodisc dimensions, small imperfections in fabrication result in large variation of the absorption wavelengths, which is why there is difference in absorption wavelengths for simulated and measured spectra.

The reflectance plots can be further analyzed to map the colors obtained from nanodiscs on standard CIE, as shown in Figs. 1(c) and 1(d) for experimental and simulated spectra, respectively. It allows for individual nanodisc colors to be marked by a single point on CIE diagram, which is important for actual color printing applications.

### 3.3. Color printing using nanodiscs

The feasibility of color prints obtained by implementing a higher-order resonant nanodisc strategy is demonstrated by a print of the logo of the University of Southern Denmark which has been modified with colored blocks to demonstrate the color quality, resolution and visual appeal of the wide range of colors. The color of each pixel is encoded by a specific nanodisc diameter corresponding to the designed color with the period of 450 nm using a computer-generated script. Five different colors of red, orange, light orange, light green and brown are chosen for the color blocks along with realistic colors for the apple, twig and leaves from the color palette of 40 nm SiO$_2$ thickness. The fabrication is described in detail in the Methods section. An optical image of the fabricated logo imaged using low numerical aperture objective to capture the whole area of $350 \times 125 \mu m$ is shown in Fig. 4(a). As seen from the optical image in Fig. 1(a), individual colors appear bright and well-saturated against the background, especially the colors of the apple designed from large size nanodiscs supporting a third-order GSPR appear realistic with a high degree of color saturation. To assess the quality of the nanodiscs, the SEM images are shown in Figs. 4(b)-4(e). The Figs. 4(b) and 4(c) show SEM image of color blocks on letter S and U with corresponding high-resolution SEM image of the outlined region in letter U in Fig. 4(d), which shows that nanodiscs with large variation in diameter are reproduced accurately.
4(e) shows an SEM image of the large diameter nanodiscs corresponding to third-order GSPR used for vibrant red and green colors shown in the optical image. Thus, uniform fabrication with stable and reproducible patterns is achieved considering the range of nanodiscs from small 75 nm to large 400 nm diameters. Most of the discs sizes investigated in this work fall within the fabrication capabilities of common nano-imprint lithography (NIL) facilities [46–49] and all discs sizes lie within the capabilities of state-of-the-art NIL fabrication [50,51].

4. Conclusion

In conclusion, the approach of higher-order GSP resonances based meta-elements to enhance many features suitable for subwavelength color printing is explored. The appropriately scaled sizes demonstrated based on third-order gap plasmon resonances significantly improve compatibility with large-scale fabrication techniques, with an obvious expansion in the range of colors as well as high degree of color saturation. The size range achieved is larger than the current sizes (<100 nm) for fundamental resonances in the visible and smaller than the wavelength, which retains the advantages of subwavelength resolution in imaging and low bulk volumes. Another advantage of large size meta-elements is the improved absorption of resonant wavelengths, which shows enhancement in color saturation producing distinct and pure colors, a feature only observed for fundamental resonance based on small elements. Moreover, it is flexible and easily programmable to implement the higher-order based strategy of appropriately scaled sizes considering the different types of metal-insulator-metal geometries and materials being explored for plasmonic color printing. Thus, the technique has potential to be integrated with current high-resolution applications like printing security markings, as well as large area applications like printing for consumer products with the advantage of wide color range and simple design strategy.
5. Methods

This section explains the detailed fabrication procedures, optical measurement schemes and numerical simulations used in our work, through the following subsections:

5.1. Fabrication

Uniform nanodisc arrays of different diameters and the design patterns are fabricated by firstly preparing an optically thick gold layer and thin SiO$_2$ layer on commercially purchased Si wafers. The gold layer and 3 nm titanium used for adhesion are e-beam deposited whereas, radio frequency (RF) Ar$^+$-sputtering is used for SiO$_2$. To fabricate the nanodisc arrays, and assess its quality, we use JEOL type e-beam lithography (EBL) and scanning electron microscopy (SEM) system (model: JEOL JSM-6490LV) with an acceleration voltage of 30 kV, a working distance of 9 mm. The e-beam lithography is a one-step process consisting of spin coating 950 kDa Poly(methyl methacrylate) (PMMA) resist on the substrate (100 nm), after which a pre-designed pattern is e-beam written with the write-field calculated from the pattern, which is 100 µm for uniform diameter arrays and 125 µm for the design patterns of SDU logo. The area-dose is varied from 150 µC/cm$^2$ for large nanodisc diameters to 380 µC/cm$^2$ for small nanodiscs. The exposed PMMA film is developed with a 3-to-1 mixture 2-propanol/methyl isobutyl ketone (MIBK) and rinsed with 2-propanol (IPA). 50 nm of gold is subsequently evaporated onto the sample and the PMMA mask is removed using acetone to obtain the nanodisc patterns. The SEM images presented in Fig. 4 are obtained with a TESCAN Mira 3 SEM at a working distance of 4 mm, using in-lens secondary electron imaging.

5.2. Optical measurements

Both optical images and spectral measurements of the nanodisc arrays are performed using a bright-field reflective optical microscope (Zeiss) for visible light (400 to 780 nm) using a tungsten halogen lamp as the light source. The sample is illuminated through 50x objective (NA 0.8) with a closed aperture stop to ensure normal incidence. The reflected light is collected by the objective and directed to a fiber-coupled spectrometer with a pinhole to collect the spectra of the selected nanodisc arrays. The spectrometer (Ocean Optics QEPro) is sensitive within the visible and near-infrared range. The reflectivity of nanodisc arrays is calculated by normalizing the reflected spectra from the nanodiscs to a silver mirror. The optical images of design patterns are imaged using a CCD camera with a 20x objective.

5.3. Numerical simulations

All simulations are performed using commercial finite element software Comsol Multiphysics (version 5.2). Single unit cell consisting of the configuration described in the paper is used as a model. Plane-wave of linearly polarized light is considered for all simulations. The nanodisc and bottom layer are comprised of gold with permittivity values taken from Johnson and Christy database, [44] the substrate comprised of SiO$_2$ is assumed to have refractive index of 1.45, while the medium above nanodiscs is chosen to be air. Perfectly matched layers are used to truncate the air domain to reduce reflections and perfect electric conductor is applied to the bottom side of thick gold layer. The edges of the nanodiscs were rounded by 5 nm. The reflected spectra above the nanodiscs are normalized directly to the incident plane wave without considering the effects such as surface roughness. For this calculation, the imaginary part of the dielectric function of gold is multiplied three times to account for grain boundary effects of gold nanodiscs in simulations.
Appendix

In this section we provide the data to support all information presented within the manuscript. This includes the electromagnetic modes corresponding to first and third order resonances for different thickness of SiO$_2$ layer (20 and 50 nm). The experimental and simulated reflectance spectra for all nanodisc diameters from 50 to 425 nm on SiO$_2$ layer of 40 nm is shown. We also compare the reflectance of nanodiscs for different SiO$_2$ layers 20 and 50 nm thick, the simulated spectra for nanodisc diameters of 75, 100 and 325 nm are shown. The data is respectively shown in graphs below:

Fig. 5. GSP mode map for different SiO$_2$ thicknesses. (a) Electromagnetic fields of first order GSP mode for D 75 nm nanodisc and third-order GSP mode for D 325 nm nanodisc for SiO$_2$ thickness of 20 nm and (b) 50 nm.

Fig. 6. Experimentally measured reflectance for all nanodiscs diameters. The spectra of nanodiscs collected using ×50 objective is normalized with silver mirror as reference.
Fig. 7. Simulated reflectance corresponding to above measured nanodiscs. 
The spectra are simulated by considering losses through three times increase
in the imaginary part of dielectric function of gold.

Fig. 8. Simulated reflectance spectra for D 75, 100 and 325 nm nanodiscs on
SiO₂ thickness of (a) 20 and (b) 50 nm.

Finally, it is important for color printing applications to know if the larger array period chosen
in our study affects the far-field spectra due to diffraction effects. To confirm that this has no
significant effect in the far-field, the reflectance measurements carried out for uniform array (D
300 nm) using two different numerical objectives is shown in Fig. 9, below:
Fig. 9. Measured reflectance spectra of an array of nanodiscs with $D = 300$ nm on 40 nm of SiO$_2$, using objectives with numerical apertures of 0.95 (50×) and 0.50 (20×), where the 50× objective collects the first diffraction of the normally incident illumination and 20× objective does not. It emerges that the collection of the first diffraction order does not alter the observed spectrum significantly.

**Funding**

European Research Council (341054, PLAQNAP); VILLUM Foundation (16498); University of Southern Denmark (SDU2020).

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