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Micropillar Cavities containing PMMA & Red-F Fluorescent Molecular Dye using Nb_2O_5/SiO_2 DBRs

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ABSTRACT

Photoluminescence emission from weakly coupled molecules has been investigated by placing a thin organic semiconductor film in one dimensional micropillar microcavities. The structure consisted of a Poly(methyl methacrylate) PMMA and red-emitting organic semiconductor (Red-F) thin film sandwiched between pair layers of dielectric mirrors made of materials that have low and high refractive indexes such as Nb_2O_5/SiO_2 respectively. The structure's different diameter micropillars were designed using a focused ion beam. This construction is able to reserve the light in the micropillars in three directions due to total interior reflection horizontally and disseminated Bragg reflectors vertically. Optical emission properties such as changing the spectral wavelength of the released light depending on the micropillars diameter can be controlled. As a result, series of sharp lines of emission spectra were obtained from the micropillars with diameters starting from 4 μm to 10 μm . By placing a 200 nm of thin film using (Red-F & PMMA) polymers into a 7 μm diameter micropillar, a quality factor of 446.1 was obtained. Besides, it was also obvious that as the micropillar diameter was decreased, the energy of all cavity modes gradually blue-shifted.

تجاويف دعامات دقيقة تحتوي على الصبغات الجزيئية PMMA و Red-F المتألقة باستخدام

Nb_2O_5/SiO_2 كعكاسات براغ الموزعة

فالح لفته مطر الجشعمي ، سحر ناجي رشيد

قسم الفيزياء ، كلية العلوم ، جامعة تكريت ، تكريت ، العراق

الملخص

تمت دراسة التالى الضوئي المنبعث من جزيئات ضعيفة الاقتران من خلال وضع غشاء رقيق من مادة عضوية من أشباه الموصلات في تجاويف دقيقة ذات شكل دعامات بعيد واحد. يتكون هيكل هذه الدعامات من غشاء بولي ميثيل ميثاكريلات (PMMA) وطبقة رقيقة من شبة موصل عضوي باعث للون الأحمر (Red-F) العضويين محصورين بين طبقات مزدوجة من المرايا العازلة المصنوعة من مواد ذات معاملات انكسار منخفضة وعالية مثل Nb_2O_5/SiO_2 على التوالي. تم تصميم هياكل الدعامات الدقيقة بأقطار مختلفة باستخدام شعاع أيوني مركز. هذا الهيكل قادر على حجز الضوء في الدعامات الدقيقة في ثلاثة اتجاهات بسبب الانعكاس الداخلي الكلي أفقيًا وعكاسات براغ المنتشرة عموديًا. يمكن التحكم في خصائص الانبعاث البصري مثل تغيير الطول الموجي الطيفي للضوء المتحرر اعتمادًا على قطر الدعامة الدقيقة. ونتيجة لذلك، تم الحصول على سلسلة من الخطوط الحادة لأطياف الانبعاث من الدعامات الدقيقة بأقطار تتراوح من 4 ميكرومتر إلى 10 ميكرومتر. من خلال وضع 200

نانومتر من الأغشية الرقيقة باستخدام بوليمرات (Red-F & PMMA) في هيكل دعامة ميكروية يبلغ قطرها 7 ميكرومتر، تم الحصول على عامل جودة قدره 446.1. بالإضافة إلى ذلك، كان من الواضح أيضًا أنه مع انخفاض أقطار هياكل الدعامة المايكروية، تتحرك الأطوال الموجية الأساسية للطاقة تدريجيًا إلى اللون الأزرق.

Introduction

A wide range of studies of the one dimensional photonic crystal have been made using a different kind of one dimensional photonic crystal such as a small diameter micropillar and vertical cavity surface emitting laser. Micropillar microcavities are appropriate structures to confine light in three dimensions due to two dimensional total internal reflections from both sidewalls of the pillars horizontally and one dimensional vertically depending on distributed Bragg reflector mirrors [1-9]. Micropillar microcavities can be defined as structures consisting of two high- low refractive indices of placed of distributed Bragg reflectors (DBRs) in both sides of an effective dipole emitter. Then, this construction is drilled perpendicularly to design a micropillar. These kinds of sources of light are expected to have applications in both of quantum-computation and quantum- cryptography systems. Furthermore, some fundamental processes like light-matter coupling and interaction can be studied using the distributed Bragg reflector Micropillars [10-16]. To get planar microcavities at high-low reflectivity, it can be used a design with N alternating quarter-wavelength thick pairs in the bottom and top of the cavity to create the distributed Bragg reflectors (DBRs) [17-18]. Electroluminescent devices which depend on thin films of organic materials receive much attention because of the need for light weight, low power colour displays and back lights. Using microcavities to modify the electroluminescence characteristics of organic materials has been researched by several groups [19]. Planar microcavities of semiconductor are able to reinforce and control the interaction between both electronic excitations and light. As a result, weak and strong coupling regimes occur when the mode of microcavity is in resonance with the excitonic transition [20]. The first arrays of "Vertical Cavity Surface-emitting Laser was created by Iga et al. in 1970s". They started with GaAs Dielectric mirror-coated thin films of a few micrometers thickness [21]. Then, in 1979 Iga et al. obtained the first lasing operation of "a GaInAsP/InP SE laser" [22]. Weisbuch et al. in 1992 studied powerful coupling phenomena in the quantum microcavity [23]. In 1993, Tsutsui et al. were the first to study the emission created by electric pumping from organic dye film-coated microcavities; then in 1994, they fabricated an optical microcavity structure composed of SiO₂/TiO₂ dielectric reflectors made of bilayers structure and organic three-layer electroluminescence [24]. In the same year, Dodabalapur et al. produced blue, green, and red emitting devices from a one substance using a novel idea depending on combining effects of Fabry-

Perot cavity with the organic material's broad luminescence [25]. In 1997, "Dirr et al. find the strong influence of the emission of thin film position on the luminescence spectra using organic material such as Alq₃ and TTFA was sandwiched between two microcavity of planar Fabry-Perot structures" [26 -27]. Lidzey et al. were the first to observe the robust coupling regime in a microcavity of organic optical semiconductors in 1998 using of organic semiconductors in microcavities [28]. One year later 1999, Tokitoa et al. created "a planar microcavity with three teams of SiO₂/TiO₂ dielectric mirrors and a top metal layer that acts as a mirror with organic material" [29]. Lin et al. investigated "the optical properties of microcavity organic light-emissive devices with two metal mirrors in 2005. The study found that the back mirror has a high reflection and the exit mirror has a low-loss high-reflection, as well as a 24 nm thin Ag mirror, which was required to improve the luminance in microcavity devices". [30]. Wenus et al. in 2006 generated the first states of hybrid organic-inorganic polariton. The structure made of twelve pairs of two similar dielectric SiN/SiO₂ DBRs mirrors were laminated together to design an optical microcavity with quality factor 200 [31]. In 2017, Dusel et al. applied thermal fingerprint technology to build microcavities of the three-dimensional pillars where light can be confined by the forming of 0D cavity mode patterns. This approach is able to form pillar geometries hemispherical rather than pillars cylindrical. Hemispherical pillars are impressed directly on the dielectric mirrors' top and then the pillars are capped by a gold thin layer [32].

Method

The structure of the micropillar microcavity is illustrated schematically in Fig. 1(a). The construction is made of a glass base on which six and a half alternative pairs of Nb₂O₅/SiO₂ quarter-wave layers were deposited. "The dielectric DBR mirrors with thickness $\lambda/4n$ were deposited using electric beam vapour deposition. Then, a thin film of active organic materials was spin-cast on the top of this DBR to make a $\lambda/2n$ layer of a Red-F molecular dye at a concentration of 10 mg with 1ml of 1-2Dichlorobenzene and at a concentration of 50 mg with 1ml of 1-2Dichlorobenzene PMMA"[7]. Fig. 1(b,c) depicts the chemical structure of the organic materials. A second DBR, eight pairs and a half of Nb₂O₅/SiO₂ quarter-wave layers, then at room temperature, it was applied to the organic material top. This design created a stop band with a width of approximately $\lambda\Delta n/n$ hence Δn is the difference of index between the layers of DBR that form the stack,

λ represents the stop band central wavelength, and n represents the index average. The structure

reflectivity is illustrated in Fig. 1(d).

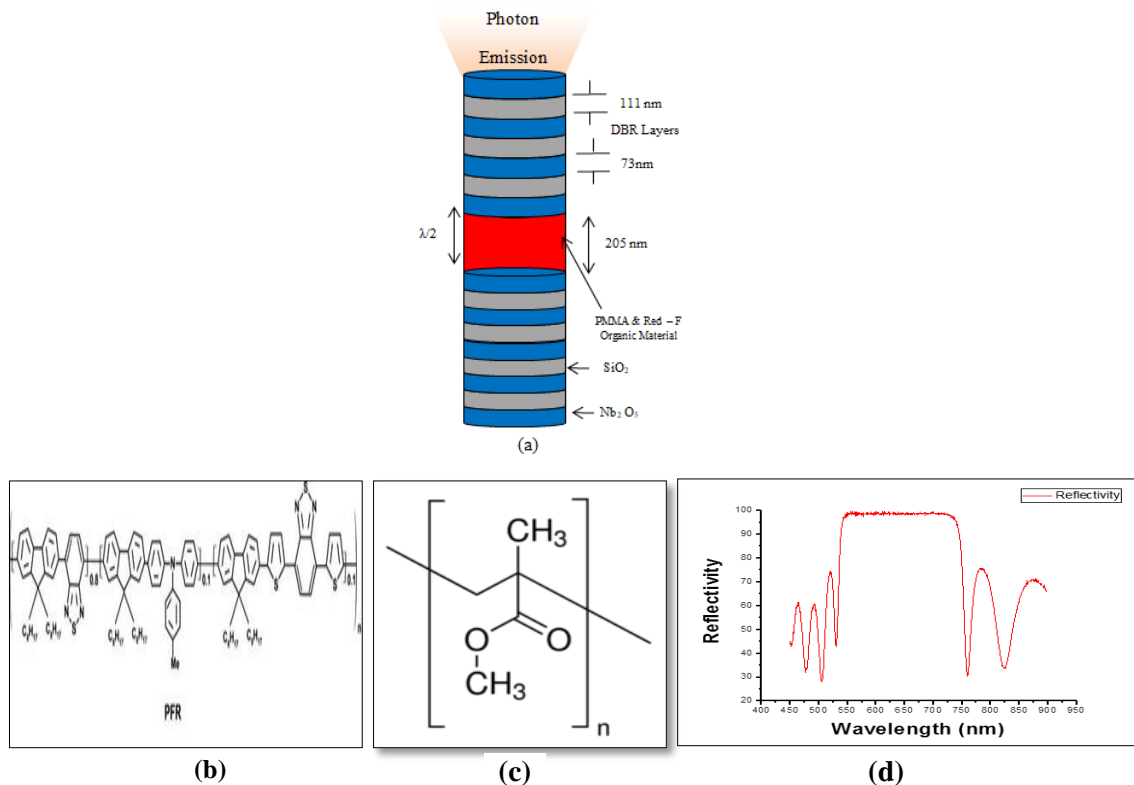


Fig. 1. shows the following: (a) A micropillar cavity schematic with embedded organic material in the center. (b) The chemical structure of the Red- F. (c) The chemical structure of PMMA. (d) The reflectivity of the DBR.

The quarter wavelength condition must be applied accurately to obtain a high reflectivity in the DBR stop band. The DBR reflectivity (r_{DBR}) for a large number of pairs, N , can be calculated as:

$$r_{DBR}(E) = \sqrt{R} \text{EXP}(i\phi_{DBR}(E)) \dots 1$$

And the reflectivity at the stopband centre, R , is given by:

$$R = 1 - 4 \frac{n_{EXT}}{n_{cav}} \left(\frac{n_L}{n_H} \right)^{2N} \dots 2$$

where n_H , n_L , are the indices of refraction for the high and low DBR refractive index at λ_0 . n_{EXT} and n_{cav} are the medium of the cavity outside (glass or air), and the material situated in refractive index of the cavity, respectively. Identical phase ϕ_{DBR} that depends approximately and linearly on the energy of photon at the natural incidence is given by:

$$\phi_{DBR}(E) = \frac{n_{cav} L_{DBR}}{hc} (E - E_s) \dots 3$$

Where E_s is the stop band central energy which is equal to $(E_s = 2\pi c \hbar / n_{cav} \lambda_s)$ where c represents the speed of light in a vacuum, λ_s is the DBR central wavelength in the cavity) and L_{DBR} is the mirror penetration depth which can be calculated as:

$$L_{DBR} = \frac{\lambda_s}{2n_{cav}} \frac{n_L n_H}{n_H - n_L} \dots 4$$

The centre of the dielectric mirrors stop band was designed to be 650 nm. The dielectric mirrors'

reflectivities peak were fabricated to be about 99% in our DBR from a classical calculation by using the refractive indexes [33-34]. But it was achieved experimentally (97%).

Photoluminescence (PL) emission from a thin film of Red- F organic material illustrated as in figure 2(a). Then, a range of micropillar microcavities, a circular trench into the planar cavity, which have a diameter between 4 and 10 μm and depth 10 μm was fabricated with a focused-ion-beam writer from a planar cavity. Figure 2(b) shows a microscopy image of those micropillars; and the properties for each micropillar are demonstrated in table 1. Figure 2 (c) illustrates an image of the scanning electron microscopy SEM of a 7 μm micropillar diameter.

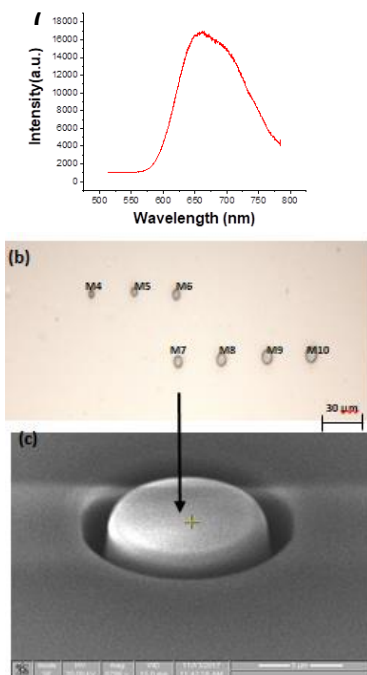


Fig. 2: Shows (a) The photoluminescence (PL) emission from a thin film of Red- F. (b) microscopy image of the micropillars.(c) scanning electron microscopy image of 7 μm diameter micropillar.

Results and discussion

We studied creating micropillars by using far field optical spectroscopy. For the measurements of far-field, the semiconductor laser (405 nm) was focused onto the surface of sample in 10 μm diameter spot. Then the micropillars PL was composed out of a 0.7 numerical aperture lens and directed towards a 0.25 m spectrometer of nitrogen-cooled charge-coupled device (CCD) that have a spectral resolution of 0.5 Å. I confirm that all the results shown here were done in air and at room temperature.

Table1. shows the properties of each micropillar

The Micropillar No.	The Diameter of the Micropillar (D) (μm)	The Depth of the Micropillar (Z) (μm)
M-4	4	10
M-5	5	10
M-6	6	10
M-7	7	10
M-8	8	10
M-9	9	10
M-10	10	10

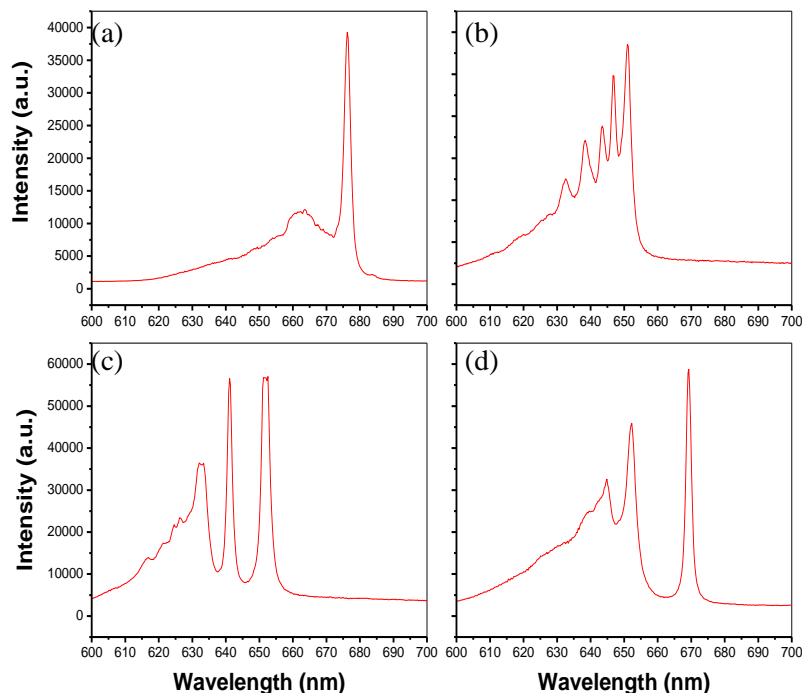


Fig. 3: Shows (a) the photoluminescence (PL) emission of the cavity fundamental mode. (b, c and d) is the micropillars emission spectra having diameters of 7, 8, and 10 μm respectively

The emission series spectra (the distinguishing fingerprint of WGM) from micropillars can be explained depending on Whispering-gallery mode phenomena, “which are basically total internal reflection (TIR) traps closed circular waves inside an axially symmetric dielectric body for a long time ” [35-43]. The optical mode spectrum in these

structures is split into a set of discrete states due to the three-dimensional confinement as given by"[7-37] eq. 5

$$E_{n_x, n_y} = \sqrt{E_0^2 + \frac{\hbar^2 c^2}{\epsilon} (k_{x, n_x}^2 + k_{y, n_y}^2)} \quad \dots 5$$

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$E_0 = \hbar c k_0 / \sqrt{\epsilon}$ represents the unetched cavity energy and k_0 is the identical wave vector. In the case of a perfectly reflecting circular cavity with sidewalls, the optical modes energies in figure 3 can be measured as

$$E_{n_x, n_y} = \sqrt{E_0^2 + \frac{\hbar^2 c^2}{\epsilon} \left(\frac{x^2 n_x^2 + y^2 n_y^2}{R^2} \right)} \dots 6$$

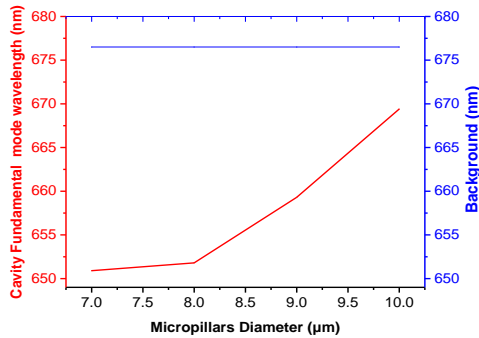


Fig. 4: represents the relation between the fundamental mode wavelengths of cavity with the micropillar diameter.

E_0 here represents the fundamental cavity mode energy, R represents the pillar radius, and $x_{n_x,0}, n_y$ is the Bessel function n_r th zero [38] "that can describe the separation of the wave equation in cylindrical or polar coordinates as a result of using the zero-order function to solve the problem of an oscillating chain suspended at one end. The relation between the fundamental mode energy and the radius pillar is illustrated in Figure 5. It is clear that the energy of the fundamental mode was decreased as the micropillars radius increased" [7-39].

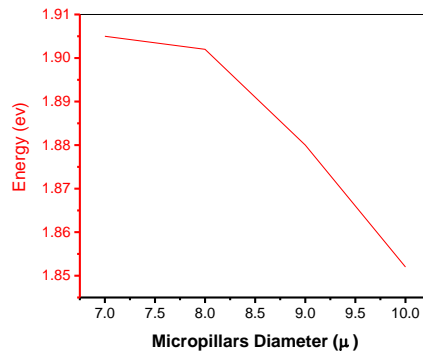
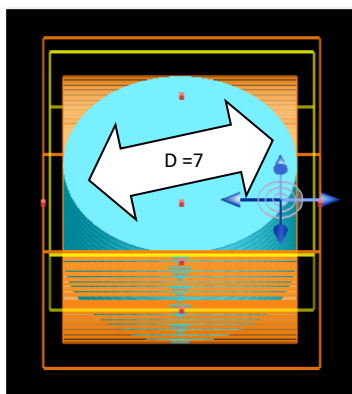


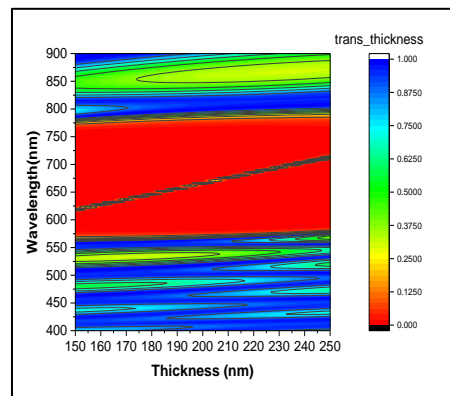
Fig.5. shows the relation between the fundamental mode energy for the microcavities as a function of the diameter which has designed

Lumerical FDTD simulations

To obtain a high accurate description for the resonances of Whispering-gallery mode phenomena in micropillars, modelling method was used. The utmost commonly used method for simulating optical devices [40] is the finite difference time domain (FDTD) Lumerical simulation process, which is used to foretell and analyze the micropillar structures emission spectra series [41-44]. The easiest way to estimate the cavity quality factor is to simulate the cavities response to the light [42]. In this study a typical micropillar cavity consisting of Nb_2O_5 and SiO_2 stack has been design to the simulation with Lumerical FDTD program, as in Fig 6 (a). The Organic material thickness used in the designing was calculated with the software of the transfer matrix as it clear in Fig .6 (b). This This mentioned program program is useful for measuring the suitable thickness of the organic material (195 nm) as a function of the required wavelength (660.4 nm).



(a)



(b)

Fig.6. (a) shows a Schematic of a micropillar cavity and an organic material was embedded in the centre by using Lumerical FDTD program. (b) the thickness of the used organic material

The positions of the Whispering-gallery mode peak were form to be conformable with the results from the experimental part as illustrated in Fig. 7 (a). The

map of amplitude for the Whispering-gallery mode peak of Fig. 7 (a) at the wavelength at 650.9 nm, 1.905 eV, is shown in Fig.7 (b).

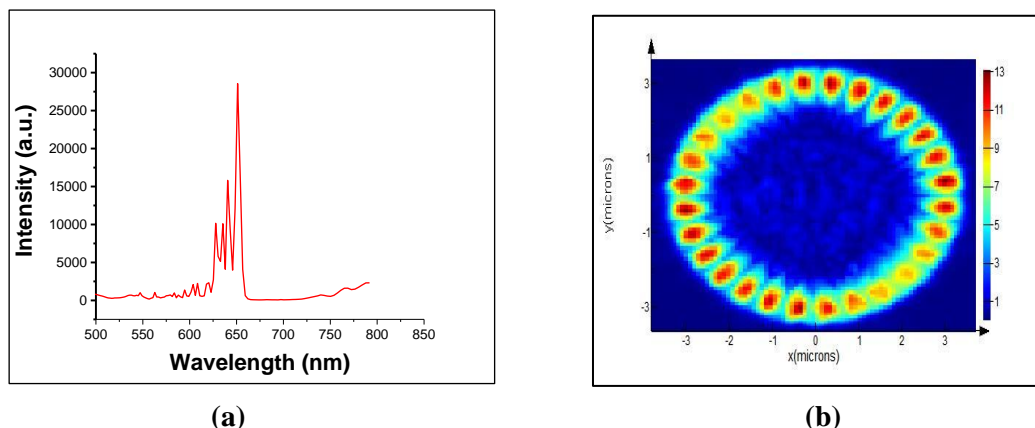


Fig.7 (a) shows the spectra emission from micropillars with a diameter 7 μm by using Lumerical FDTD program. (b) The amplitude map for the Whispering-gallery mode peak at 1.905 eV

Conclusion

We have created micropillar microcavity structures. The photoluminescence emissions from weakly coupled by placing a Red-F organic material thin film in a one dimensional micropillar microcavity structure have been investigated. As a consequence, Q-factor of 446 was obtained by placing the polymer into a 7 μm micropillar diameter. We observed that properties of optical emission such as changing the emitted light spectral wavelength depending on the micropillars diameter could be controlled. Besides, it is as well obvious that all cavity modes energy gradual blue-shift happens when the diameter of the

pillar is reduced. Series of emissions consider as a characteristic imprint of whispering gallery mode spectra because of the smooth sidewalls of the micropillars. The lumerical FDTD program has been used to stimulate the empirical results and obtained a good agreement.

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