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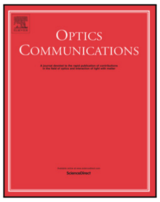

# Fabrication and infrared-transmission properties of a free-standing monolayer of hexagonal-close-packed dielectric microspheres

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ABSTRACT

This paper presents a self-supporting technique for fabricating a free-standing monolayer (FSM) of hexagonal-close-packed (HCP) polystyrene (PS) microspheres, based on a combination of Langmuir–Blodgett method and a robust transferring technique. Three narrow bandwidth resonances (NBRs) with quality factors as high as 142 are experimentally observed for the first time in the transmission spectra of the FSM of 1.0-μm-diameter PS spheres. Further numerical simulations confirm that these NBRs supported by the FSM originate from the excitations of photonic eigenmodes of the array. In addition, these NBRs are experimentally demonstrated to be highly tunable by simply changing the size of PS microsphere. Moreover, we further demonstrate both by experimentally and theoretically that the incorporation of a dielectric substrate to the monolayer of HCP dielectric microspheres have adverse effect on obtaining high-*Q* resonances compared to the free-standing case, i.e., broaden the linewidths and decrease the amplitudes of the observed resonances.

## 1. Introduction

When monodisperse dielectric microspheres are arranged into the two-dimensional (2D) array, it is not known as the 2D colloidal crystals (CCs), but also used as a powerful and low-cost promising approach to fabricate versatile metal or dielectric micro/nanostructures. For example, the monolayer of close-packed colloidal crystals have been used as templates for the preparation of planar metal nanostructures [1–4], the quasi-three-dimensional metallic half-shells [5–7], through-pore or nanorod array [8,9], and even the growth of binary colloidal crystals [10,11]. In addition to obtaining the various nanostructures based on the nanosphere lithography technique [12–14], monolayer of hexagonal-close-packed (HCP) or square lattice of dielectric microspheres itself especially for the free-standing case [15,16], has been theoretically demonstrated to support high-*Q* photonic eigenmodes (PEMs) with the ability to efficiently confine light within or in the surroundings of the dielectric microspheres, which may find important applications in 2D distributed feedback lasers and 2D corrugated waveguides [17,18]. However, to the best of our knowledge, the free-standing monolayer (FSM) of submicron dielectric spheres and the corresponding high-*Q* PEMs have not been experimentally reported in the literature. Among various self-assembly techniques for preparing monolayer of close-packed dielectric microspheres, including spin coating [19], solvent evaporation [20–22] and interfacial-assembly [23], the Langmuir–Blodgett method have gained much attention because it can naturally transfer the monolayer of dielectric microspheres formed on the water/air interface onto many types of substrates [24,25], which may be one of the most promising approach to achieving the FSM of dielectric microspheres.

In this letter, we report a robust method to prepare a free-standing monolayer of HCP polystyrene (PS) microspheres by transferring the monolayer formed on the water/air surface onto the substrates with the through-holes. Three narrow bandwidth resonances with maximum quality (*Q*) factor of 142 are observed experimentally in the transmission spectrum of the FSM of 1.0-μm-diameter PS spheres. Their physical origins as a result of excitations of PEMs of the array are further confirmed by our numerical simulations. In addition, we demonstrate that these high-*Q* PEMs are not only highly tunable, but also extremely sensitive to the refractive indexes of dielectric substrates.

## 2. Sample fabrication and characterization

The process for the preparation of FSM of PS microspheres is based on our newly developed self-supporting technique. In brief, a monolayer

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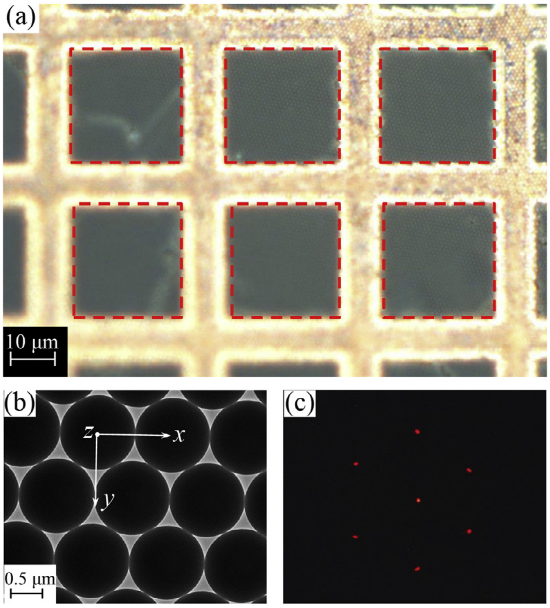

**Fig. 1.** (Color online) (a) Optical microscopy image of a monolayer of HCP PS microspheres (1.0-μm-diameter) self-suspended onto a substrate with through-holes. The red dashed-line boxes indicate the free-standing areas. (b) Typical TEM image of the FSM showing the perfect HCP arrangement of PS microspheres. (c) Diffraction pattern of the as-prepared FSM using the laser of 633 nm.

of monodisperse PS spheres with diameters of 1.0, 1.1, 1.3 and 1.6 μm (coefficient of variation less than 3%, purchased from Thermo-Scientific) is first self-assembled on the water–air interface using a modified Langmuir–Blodgett method [26] and then transferred onto a grid substrate (copper mesh with diameter of 3 mm) with hundreds of micrometer-sized through-holes to form self-suspended monolayer by slowly draining the water for about two hours. The resulting self-suspended PS microspheres array located within the through-holes can thus act as the FSM. Fig. 1(a) shows the optical microscopy image of the as-prepared FSM with PS diameter of 1.0 μm. It is directly seen from Fig. 1(a) that the large-area FSM of PS microspheres (indicated by the red dashed boxes) is formed within the through holes due to the strong interparticle van der Walls interactions and the interactions between the particles and the grid substrates. In Fig. 1(b), a typical transmission electron microscopy (TEM) image clearly confirms the nearly prefect hexagonal-close-packed arrangement of PS microspheres in the FSM. In addition, the long-rang-order of the as-prepared FSM is further verified by the laser ($\lambda = 633$ nm, spot size: 1 mm) diffraction pattern shown in Fig. 1(c).

## 3. Results and discussion

The near-infrared transmission spectra in this paper are measured with a commercial Fourier-transform infrared spectrometer (Nicolet 5700) equipped with a linear polarizer. The quasi-collimated light (diameter: 50 mm) is focused by an off-axis parabolic mirror (focus length: $\sim$250 mm) onto the sample, where the focused light with a spot diameter of $\sim$ 2 mm has a divergence angle of $11^{\circ}$. The incident light polarization configuration in this study with respect to the ordered array of PS microspheres is schematically shown in Fig. 1(b). The coordinate system is chosen to make sure that the PS microspheres lie on the $xy$-plane. Under normal incidence, the measured transmission spectra of two orthogonal polarizations ($x$-polar. and $y$-polar.) show no difference in spectral features due to the spherical symmetry in the $xy$-plane of our sample (see Figure S1(a)). So, for simplicity, the measured transmission spectrum of the FSM of PS microspheres (1.0-μm-diameter) with only one polarization ($x$-polar.) is displayed in Fig. 2(a) (black trace). It is directly seen from Fig. 2(a) that three narrow bandwidth resonances (NBRs) are observed in the transmission spectrum, which is located at 1199 nm (A1), 1190 nm (A2) and 1006 nm (A3), respectively.

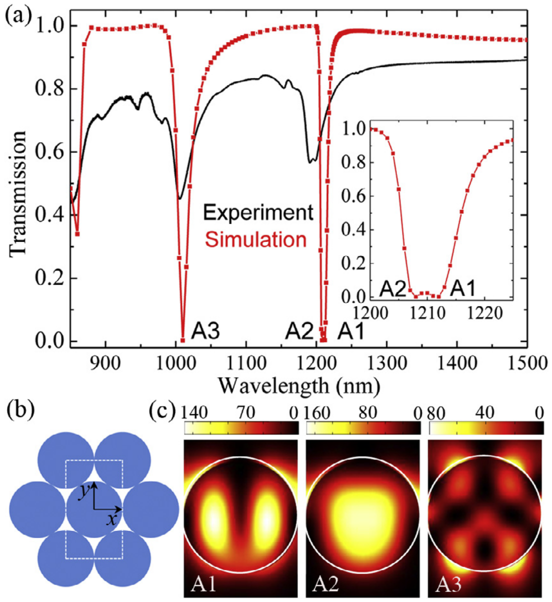

**Fig. 2.** (a) Measured (black trace) and simulated (red trace) transmission spectra of a FSM of HCP PS spheres with diameter of 1.0 μm. An enlarged simulated spectrum of A1 and A2 modes is shown in the inset. (b) Top-down view of the FSM. The dashed box indicates the calculation domain, consisting of one complete and four quarter dielectric microspheres. (c) Total electric field intensity spatial distributions ($xz$-plane) of A1, A2 and A3 modes in the FSM at the wavelengths of 1212, 1208 and 1010 nm, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

In order to understand these observed NBRs supported by the FSM, numerical simulations are carried out using the three-dimensional finite-element-method [27,28]. The simulated unit cell, shown as the dashed rectangle in Fig. 2(b), consists one complete and four separate one-quarter of the dielectric microspheres. In the simulation, its four sides are applied with periodic boundary conditions. The refractive index of PS microspheres is taken to be 1.59. Similar to the experimental results, the simulations of two orthogonal polarizations ($x$-polar. and $y$-polar.) present the similar spectral features (see Figure S1(b)). So, the simulated transmission spectrum of the FSM of dielectric microspheres (1.0-μm-diameter) with polarization along $x$-axis is shown in Fig. 2(a) (red trace). The inset shows an enlarged simulated transmission spectrum around 1210 nm, which clearly displays two discrete A1 and A2 modes. From Fig. 2(a), it is directly seen that the simulated transmission spectrum (red trace) presents a good agreement with our measurements (black trace). The remaining discrepancies between measurements and simulations, such as little resonance wavelengths blue-shifting and transmission dips broadening in the measurements comparing with simulations, most probably because of the weak dispersion of PS materials and the fabrication tolerances in experiment. In addition, our simulations can also reveal the spatial distributions of electric field intensity for the observed three NBRs. Fig. 2(c) shows the field intensity distributions ($xz$-plane) associated with the A1–A3 modes at the wavelengths of 1212, 1208 and 1010 nm, respectively, revealing a strong concentration and large enhancement of electromagnetic field within (A1 and A2 modes) or in the surroundings (A3 mode) of the dielectric microspheres. These field distribution features reveal that a FSM is able to effectively confine light through the excitations of PEMs of the coupled array arising as a consequence of multiple scattering by individual microsphere [16].