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Mechanical and Electric Control of Photonic Modes in Random Dielectrics

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Random dielectrics defines a class of non-absorbing materials where the index of refraction is randomly arranged in space. Whenever the transport mean free path is sufficiently small, light can be confined in modes with very small volume. Random photonic modes have been investigated for their basic physical insights, such as Anderson localization, and recently several applications have been envisioned in the field of renewable energies, telecommunications, and quantum electrodynamics. An advantage for optoelectronics and quantum source integration offered by random systems is their high density of photonic modes, which span a large range of spectral resonances and spatial distributions, thus increasing the probability to match randomly distributed emitters. Conversely, the main disadvantage is the lack of deterministic engineering of one or more of the many random photonic modes achieved. This issue is solved by demonstrating the capability to electrically and mechanically control the random modes at telecom wavelengths in a 2D double membrane system. Very large and reversible mode tuning (up to 50 nm), both toward shorter or longer wavelength, is obtained for random modes with modal volumes of the order of few tens of $(\lambda/n)^3$.

Random dielectrics have attracted a lot of interest in the past decades for the fascinating, complex and elusive physics of disordered media, but also for a wealth of interesting applications ranging from sensors to quantum electrodynamics effects. Within transport models, light propagation in random dielectrics is described in terms of multiple elastic scattering and light coherence plays a relevant role with large deviations with respect to usual diffusion. In particular, if the degree of disorder is high enough, that is when the transport mean free path is comparable with the wavelength, electromagnetic waves can interfere in a complex manner leading to light localization.^[1,2] This is usually defined as strong or Anderson localization, where disorder causes electromagnetic transport to a halt, generating randomly shaped standing wave patterns in which light is trapped.^[3] The introduction of optical gain in random structures

may lead to stimulated emission of multiple scattered waves and then to the largely studied random lasers,^[4–6] recently also proposed as efficient sensors.^[7] The understanding of light propagation through scattering media has led to the ability to image through turbid media,^[8,9] to focus light by scattering^[10,11] and to spectrally resolve light.^[12] Light trapping can also be used to improve the absorption efficiency of thin film solar cells whenever the external surface would include a random dielectric layer.^[13] Finally, in quantum cavity electrodynamics, the Purcell effect (or even the strong coupling) of a quantum emitter in resonance with a random mode has been considered to bypass the bottlenecks of both relatively large footprint of the photonic structure and of precise spatial location of the emitters in case of photonic crystal nanocavity.^[14] For this proposal, the deterministic control of the employed random mode is of the utmost relevance due to additional disorder introduced by the fabrication process, which results in unavoidable deviations with respect to the designed structure. In particular, a spectral tuning method that preserves all the other features of the mode is required to control the exciton–photon coupling. Recently, a first step in this direction has been demonstrated by performing nano-oxidation of the dielectric membrane that hosts the disordered medium, in order to achieve few nm blue spectral shift

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Figure 1. a) Scanning electron microscopy image showing the upper freestanding microbridge, in planar view. The bottom panel is a cross view sketch with the definition of the parameters h and d_0 . b) Typical PL enhancement spectrum at a given tip position. Inset: histogram of the number of modes for different spectral ranges. c) Spectral and spatial detail of a random mode. Inset: map of the random mode. d) Maps of the maximum PL enhancement in six spectral regions. All the white scale bars in the figures correspond to 1 μm .

of an individual random mode.^[15] Still this approach is not reversible, it induces a mode shift only to shorter wavelengths, it cannot be used on a large scale, and eventually it can produce intrinsic (Q reduction) and extrinsic (emitters quenching) optical degradation of the sample. Here we achieve a reversible, fine but very large spectral tuning to a specific target wavelength, of all (of the order of 300) the random photonic modes supported by the device under investigation. These random modes are characterized by very small modal volumes and their spatial distribution is conserved over the whole tuning range.

The investigated system is a random dielectric structure on two GaAs parallel membranes with air holes positioned in random locations, with the only not-overlapping constraint. Both near field and conventional optical microscopy of the emission of embedded InAs quantum dots (QDs) are used to map the randomly localized photonic modes in space and frequency. By controlling the air distance between the two membranes, we induce either red or blue spectral shift, depending on vertical parity of the random modes. We demonstrate two possible approaches for mode tuning based on i) the local nanoforce exerted by the near-field tip and ii) the global electrostatic force obtained by the application of an external voltage bias across the membranes. Simultaneous and reversible wavelength tuning of all the modes is obtained with a record of 50 nm. Remarkably, the spectral tuning is achieved without changing the spatial distribution of the modes, thus addressing the request needed to match the random mode with an emitter. The random dielectric system is realized by fabricating air holes in two parallel membranes ("double-membrane," DM) with nominal thickness of $h = 180$ nm, separated by a nominal

inter-membrane distance $d_0 = 250$ nm along the growth direction. InAs QDs, whose ground state is centered at 1300 nm, are embedded in the middle of the upper membrane. **Figure 1a** shows the top view scanning electron microscopy image and a cross view sketch of the sample. These air hole patterns have previously demonstrated to support Anderson localized modes in the telecom wavelength range, which is relevant for photonic applications.^[16] The vertical etching assures that the random holes patterns in the two membranes have the same nominal design, which is of the utmost relevance for tuning experiments. The four bridges supporting the upper membrane are designed to allow a large deflection of the top membrane with the nN force exerted by the SNOM tip.^[17] The other panels in Figure 1 report data obtained by SNOM experiments. We first evaluated the spatially averaged spectrum over a large area ($7 \mu\text{m} \times 10 \mu\text{m}$) which smooths out the random modes and gives the PL spectra of the embedded QDs. Then we calculated the PL enhancement spectrum at a fixed position by dividing the recorded near field spectrum by the averaged PL spectrum. A typical example is reported in Figure 1b. Details on the experimental procedure are given in the Supporting Information. In the near field spectra collected at fixed tip positions, several sharp resonances, related to the random modes of the system, are clearly visible. In each SNOM tip collecting area, with 250 nm lateral size, we found roughly 10 random modes. These resonances are spectrally sparse over the full emission range of the QDs with quality factors ranging from 100 to 200. The inset of Figure 1b shows the number of modes collected in the central area of the DM ($7 \mu\text{m} \times 11 \mu\text{m}$) for different spectral regions. Explicitly, more than three hundred of modes, sparse

over 300 nm spectral width, were collected (see the Supporting Information). A typical spectrum of a single isolated random mode is given in Figure 1c. In the inset is reported the spatial distribution of the PL enhancement, which reflects the spatial distribution of the electric field intensity.^[18] Interestingly, the spatial extension of this mode is quite small with a mode volume similar to the one characterizing photonic crystal nanocavities (PCCs). Figure 1d displays six collective maps in which we report the maximum PL enhancement measured in the spectral regions reported on the top of each map. Note that these are neither single mode maps, nor maps at a single emission frequency. They are collective maps where each bright spot refers to the brightest mode (or sometimes even to the superposition of more modes) in that spatial region, within the selected spectral window. These maps (as well as the map in the inset of Figure 1c) are collected with the tip at a distance of the order of 10 nm to the membrane, that is, when the elastic force sustaining the upper membrane balances the force of the tip, which is fixed by the feedback loop. From these maps, we can take a census of the relevant modes and then, in case, evaluate for each mode its spatial distribution, as done in the inset of Figure 1c. We observe a large number of random modes distributed over the whole patterned area, even if different regions tend to support more efficiently modes in different spectral ranges. Another important feature of these random modes is that they can be spectrally isolated, due to their large frequency dispersion. Therefore, in a spatially resolved experiment we can deal with spectrally isolated resonances, as expected in the localized regime.^[16] Indeed, the modal volume of these random modes is few tenths of $(\lambda/n)^3$, comparable with the best PCCs, where a large surrounding photonic crystal area is needed. This shows that the areal density of random modes is almost one hundred times higher than standard PCCs.

The disorder-based device is realized by fabricating air holes in two parallel membranes (DM) with nominal thickness of $h = 180$ nm, separated by a nominal inter-membrane distance $d_0 = 250$ nm. Details of the fabrication processes are given in refs. [17,19,20]. Specifically, the selected bridge geometry is composed of a rectangular photonic area ($12\ \mu\text{m} \times 8\ \mu\text{m}$) connected to a supporting frame by four external microarms ($2\ \mu\text{m} \times 2\ \mu\text{m}$). Additional external trenches (not shown in Figure 1a) are patterned close to the supporting arms in order to release the internal stress of the structure, which causes upward buckling of the structure^[20] by an in-plane extension of the arms. Within the two membranes, two 50 nm thick layers facing each other are n and p doped (upper and lower, respectively) to realize a p-i-n diode for electromechanical control of the intermembrane distance d by means of an electrostatic force. The photonic pattern consists of air holes with 220 nm diameters, whose positions are designed by a random sequential addition generator, with an overall filling fraction of 25% and the constraint of minimum distance (1.3 hole diameters) in order to avoid hole merging.^[16] This design corresponds to the maximum practical randomness in a real sample. In other words our design defines a correlation length scale which can be measured by the presence of peaks in the structural factor (while a perfect random structures should have a flat structural factor); still our design is, within the pore nonoverlapping constraint, gives the “less structured” structural factor, justifying

the denomination throughout all the paper of random system and random modes (see discussion in ref. [2]). Electron beam lithography and reactive ion etching are used to simultaneously pattern both membranes with the same air hole pattern. Finally, a layer of high-density ($\approx 300\ \mu\text{m}^{-2}$) InAs QDs, whose ground state is centered at 1300 nm, is embedded in the middle of the upper membrane. This choice, which allows us to easily probe the random modes by the room temperature broad emission of the QDs around 1300 nm, is not decisive and different kind of emitters (as examples colloidal QDs^[21] or carbon nanotubes,^[22]) could have been used. It is also worth stressing that the QD spatial distribution is homogeneous on the scale of the spatial extension of the random modes and their absorbance is quite small and not relevant in determining the random mode Q factor (photonic crystal cavities with $Q \approx 10\,000$ can be obtained^[23]).

Due to the elastic bridges at the sides of the upper membrane, the tip force induces a deterministic reduction of the intermembrane distance d by controlling the vertical position of the sample (keeping the tip fixed) by piezo-actuators with a precision of the order of 1 nm. Assuming that the distance between the tip and the upper membrane is almost unchanged during the vertical scan (due to the SNOM feedback mechanism), the two membranes come closer when the sample is moved upward. In this way, the sample vertical position is directly related to the intermembrane distance d . The upper membrane would slightly bend with a minimum in the position of the tip^[17]; still the quote reduction is of the order of 100 nm while its lateral size is 100 times larger. This means that for the effects on the random modes within the lateral collection size of the tip (250 nm) we can safely assume a vertical rigid translation of the upper membrane without any deformation of it. The maximum reduction of the intermembrane distance depends on both the tip and sample details, which are difficult to model. In order to select the optimal tip for the compression, we performed different tests on photonic crystals cavities in a DM configuration. The best tip gives a typical intermembrane distance reduction of the order of 100 nm with a record of 140 nm.

We now address the physics of a DM device that allows the deterministic control of random photonic modes. Figure 2a shows the calculated wavelength dispersion of the first two transverse electric (TE) guided modes supported by a homogeneous (i.e., unpatterned) double membrane (thickness $h = 180$ nm) as a function of the intermembrane distance d . The symmetry of the problem leads to the splitting of symmetric (S) and antisymmetric (AS) modes. For large d ($d \geq 400$ nm), AS and S modes are degenerate. By reducing d , these modes split and shift toward opposite directions. In particular, the S mode shifts to longer wavelengths while the AS mode shifts to shorter wavelengths. When the separation d is below 100 nm, the spectral separation between these modes largely exceeds 100 nm. This feature has been used to fabricate tunable photonic crystal cavities with spectral shift up to 40 nm.^[17] Here, we show that this feature can be exploited also for the much more entangled mode distribution of a random photonic system. We performed finite-difference time-domain (FDTD) method simulations of a DM system by considering the nominal hole pattern employed for fabricating the sample

Figure 2. a) Analytical calculations for the TE S and AS modes dispersion in a homogeneous double membrane. b) FDTD calculated spectra as a function of the intermembrane distance; AS (S) resonances are filled with blue (red). c) FDTD spectra (for five different values of d) of a selected AS mode by narrow spectral excitation. d) Spectral position and quality factor (Q) of the selected mode as a function of the intermembrane distance. e) Spatial distributions ($3 \mu\text{m} \times 3 \mu\text{m}$) of the selected mode in the upper membrane for three different intermembrane distances; the white scale bar corresponds to $1 \mu\text{m}$.

in Figure 1a. Figure 2b shows the calculated FDTD spectra, collected by a $3 \mu\text{m} \times 3 \mu\text{m}$ sensor as a function of wavelength for different inter-membrane separations for both AS (blue) and S (red) modes. Large tuning of the modes is observed. In order to get insights on the origin of this tuning, we used a narrow (2 nm) spectral excitation by defining a point-like dipole located in the maximum of the electric field of a specific random mode. For every value of the intermembrane distance d , we adapt the central frequency of the employed excitation to follow the mode tuning. This allows us to selectively excite and follow the evolution of a single random mode. Figure 2c shows the calculated FDTD spectra as a function of the wavelength for different inter-membrane separations (from $d = d_0 = 250 \text{ nm}$ to $d = 50 \text{ nm}$ at 50 nm steps) of one selected AS random mode; similar results are found for S modes (not reported). Interestingly, a large shift with a minimum degradation of Q -factor is obtained (see Figure 2d). In details, a wavelength shift up to 30 nm without any reduction of Q , or more that 100 nm of spectral shift with only a 30% reduction of Q can be obtained for $d = 150$ and $d = 50 \text{ nm}$, respectively. One relevant application of spectral tuning is in view of coupling the photonic mode with localized quantum emitters (such as QDs). Here, in order to avoid the loss of spatial matching between exciton and photon, it is of the utmost relevance that the photonic hot spots do not change during the spectral tuning. Also in view of a tunable random laser, it would be important that the spatial and angular mode properties stay steady during the tuning. Then, a fundamental question, relevant for the application of random modes to Purcell enhancement, is whether the tuning occurs with minor modifications of the mode spatial profiles. Figure 2e reports the FDTD mode spatial distribution profiles of the selected random mode for three intermembrane distances. In all the points of each map of Figure 2e, the mode spectra are identical with a

tolerance of 0.1% and 10% on the spectral peak positions and on the Q factors, respectively. Up to a tuning of 60 nm, the mode spatial distribution is almost unchanged (see the Supporting Information for more details). Similar results are found for S modes (not reported).

Deterministic control of random photonic modes can almost be considered a contradiction. Even if from Maxwell equations we can solve exactly any dielectric problem, in real random samples the measured modes show large deviations with respect to the simulated ones from the nominal design due to unavoidable fabrication imperfections. As a consequence, it is almost impossible to predict where the photons localization will occur in a given sample. A key aspect of our approach is to make deterministic control the random resonances possible on top of a given unpredictable spatial distribution. This is based on the delocalization of the random modes over the two membranes, which have nominally identical hole patterns as expected by the physics of electron beam lithography and reactive ion etching steps. With this assumption, and based on the FDTD simulations of the DM system, we do expect a deterministic, reversible, fine but large spectral tuning of all random modes sustained by the DM random system. In the following, we present the experimental results obtained via the tip-induced tuning. We exploit the room temperature broad photoluminescence (PL) emission of the QDs to internally pump the random modes. The experimental apparatuses are a near-field optical microscope (SNOM) with a dielectric tip and a micro-PL setup with $50\times$ magnification and $\text{NA} = 0.45$. The continuous-wave pumping lasers are either a 780 nm diode laser or a 532 nm Nd:YAG laser. The PL signal is dispersed by a spectrometer and detected by an InGaAs array. We use illumination/collection configuration: the SNOM tip raster scans the sample surface at a fixed distance, of the order of few tens of nm, pumps the

Figure 3. a) Sketch of the DM tip compression, where we exaggerated the bending for sake of illustration. The small black arrows on the membranes borders indicate that, during compression, the quote of the sample is raised. b) Typical enhancement spectra (with many modes), reported in a color map, at different DM compression. Note the dominance of AS modes, but also the presence of one S mode on the long-wavelength side. Inset: sketch of the mode vertical distributions, taking into account the smaller thickness of the upper membrane; note that the QDs (black triangles) are only present in the top membrane. c) Detail of a well spectrally isolated AS mode with a tuning of 30 nm. d) Summary of the average tuning at $\Delta d = 80$ nm for different mode wavelengths.

QDs with pumping power in the range of 50 μ W at 780 nm and collects the QD-PL in each point at step of 100 nm with a spatial resolution of 250 nm. We first analyze the mode tuning by mechanical compression of the upper membrane due to the force applied by the SNOM tip to the upper membrane (up to 40 nN, see ref. [17]) when the quote of the sample is raised. The sketch of the DM tip compression is given in **Figure 3a**. The advantage of using a SNOM tip (with respect for instance to an atomic force microscopy (AFM) tip) is that we can mechanically modify the DM system and simultaneously probe the optical effect of this modification on all the random modes that show some spatial extension below the tip, in a region delimited by SNOM spatial resolution (250 nm). **Figure 3b** reports a map of typical spectra, collected at fixed spatial position on the upper membrane, as a function of wavelength and for different deformations of the upper membrane. More than ten modes are observed in a near field spot with diameter of 250 nm. By reducing the intermembrane distance d (from its rest position at $d_0 = 250$ nm), we observed an almost rigid blue shift of all the photonic random modes overlapping inside the detection spot of the SNOM tip. The blueshift of the random modes denotes their AS nature (see **Figure 2a**). The small reduction of the upper membrane thickness, related to the etching process,^[17] slightly breaks the vertical symmetry leading the AS (S) mode to be more localized in the upper (lower) membrane (see the inset of **Figure 3b**). Since the QDs are embedded only in the upper membrane, this allows us to select mainly AS modes and simplify the data analysis. In few cases, we were able to observe also S modes characterized by a red shift, as reported in **Figure 3b**. This demonstrates a proof of principle for tuning random modes in both directions. A different epitaxial structure can be adopted to optimize the red tuning only (i.e., thicker upper membrane) or even to obtain balanced systems with simultaneous red and blue tuning of S and AS modes (i.e., identical thickness of the two membranes). Note that the shift can be finely and reversibly controlled over a wide spectral range. Indeed a striking point of our approach is the possibility

to finely tune the mode by a fraction of nm over a tuning range of more than $\Delta\lambda = 30$ nm, as can be seen for the spectrally isolated mode reported in **Figure 3c**. We also collect the statistics of the spectral tuning for a variation of the intermembrane distance (Δd) of 80 nm as a function of the emission wavelength, which is reported in **Figure 3d**. A clear correlation exists: modes at longer wavelengths show a larger tuning, accordingly to FDTD simulation. This is likely associated to the longer extension of the vertical evanescent tails of lower energy modes. It is worth noting that the experimental spectral shift of the random modes is of the same order of magnitude of the spectral shift of the FDTD predictions, denoting that the assumption that the spatial pattern of the random modes on the two membrane is identical (employed in FDTD simulations) is verified. This also denotes the major role of vertical light confinement. Even if a quantitative comparison between simulation and experiment is not possible due to the fabrication tolerances, the agreement in the trend is good, including the fact that the tuning is larger for modes with longer wavelengths.

In order to move toward possible integration of the DM devices in a photonic chip, we explore the possibility of an electromechanical tuning of random modes by inversely biasing the p-i-n diode across the DM. A sketch of the experiment is given in **Figure 4a**, where we did not report any bending of the top membrane since the electrostatic attracting force is distributed homogeneously over the whole membrane. An important point is that the electromechanical tuning has a theoretical limit (denominated pull-in limit) for the minimum intermembrane distance d_{pi} , below which the system experiences an instability that brings the two membranes into contact. The pull-in limit states that, regardless of the choice of the design of the plates, d_{pi} is 2/3 of the original gap (i.e., that the maximum displacement of the upper membrane is $\Delta d_{pi} = d_0/3$).^[17,19] For larger absolute values of the external bias, a balance between the electrostatic and elastic forces is not possible and the upper membrane collapses on the lower one resulting in the permanent stiction of the membranes, if antistiction methods are not employed.^[24] We used micro

Figure 4. a) Sketch of the experiment. b) Typical blueshift of two random modes (the black and red triangles in panel (c)) at different external bias. c) Enhancement spectra, showing many modes, at different external bias. d) Blueshifts obtained for all the resonances of panel (c) for the maximum applied bias, $V = 3.5$ V. e) Comparison between analytical model for the electric tuning of the intermembrane distance d and values extrapolated by experimental data.

probe stations to electrically contact the sample. Since the space constraints of our SNOM setup do not fit with the probe station size, we moved to a standard micro-PL setup. This is reflected in the enhancement spectra at different external bias reported in Figure 4c, where a large spectral overlap of the random modes is observed with respect to Figure 3b due to the larger collection spot. Still, by comparing the spectra at different bias conditions, a clear blueshift of the random resonances is found, denoting the success of the electromechanical tuning, in the whole spectral range. Note also that the overall features of the spectra are maintained during the mode tuning, reflecting the steadiness of the in-plane spatial distribution of the random modes, as expected from FDTD simulations. Examples of the spectral tuning are given in Figure 4b for two modes highlighted with red and black triangles in Figure 4c. The summary of the maximum of electrical tuning for 3.5 V bias is given in Figure 4d, denoting a larger tuning at longer wavelength as observed in Figure 3d. The electrical bias can reversibly displace the two semiconductor slabs since they will return to their equilibrium position when no voltage is applied, due to the restoring elastic force. The maximum theoretical displacement of the upper membrane is given by the pull-in limit ($\Delta d_{pi} = 83$ nm). From the experimental data, we can also get a test of the electromechanical model governing the bias control of the intermembrane distance. From the experimental mode tuning by SNOM tip, we can precisely relate the mode spectral shift to the reduction of the intermembrane distance d . The comparison between the experimental data and the analytical model (see also the Supporting Information) is given in Figure 4e.

In summary, we have shown that it is possible to introduce a deterministic, reversible, mechanical or electrical post fabrication control of strongly confined random modes. More strikingly, in two-dimensional photonic structures on a DM system, we have demonstrated that one can largely and precisely tune the random resonances without altering their mode spatial profile and their quality factor. This achievement opens unpaved routes for addressing basic physics and applications in random media. As an example, spectral tuning of strongly confined

random modes can be used to tailor the radiative rate of a quantum emitter, exploiting the large areal density of random modes and overcoming the long lasting problem of spatial and frequency matching between exciton and photon. Similarly, such a control can be of the utmost relevance in random laser devices. More generally, random photonics materials, besides inducing localization of light have raised a lot of interest in their diffusive regime. Indeed, they have been proposed as an alternative and complementary platform to develop a large variety of applications in both classical and quantum realm. Our achievements, in terms of a deterministic post fabrication control of random modes, constitute an important step toward the full engineering of the random photonic platform.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

FDTD simulations, near-field spectroscopy, optomechanical devices, photonic tuning, random dielectrics

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