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Spatial steadiness of individual disorder modes upon controlled spectral tuning

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Recent innovative applications in disordered photonics would strongly benefit from the possibility to achieve spectral tuning of the individual disorder localized photonic modes without affecting their spatial distributions. Here, we design and fabricate a two-dimensional disordered photonic system, made of a GaAs slab patterned with randomly distributed circular air scattering centers, supporting localized light modes with very small modal volume. The photoluminescence of InAs quantum dots embedded in the slab is used as a probe for near field experiments and gives direct access to the electric field intensity distribution of the localized random modes. We demonstrate that laser assisted oxidation of the GaAs slab performed by near field illumination can be used for a gentle tuning of the individual random modes without modifying the subtle balance leading to light localization given by multiple scattering. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). [http://dx.doi.org/10.1063/1.4946852]
The problem of light transport and localization in disordered systems is gaining attention also for practical applications. 2D light confinement in disordered media can be used for improving the absorption in thin-film solar-cells by patterning the absorptive layer with a random distribution of holes as well as with scattering elements randomly placed on the external surface. 18 In the field of photonic based devices, the insertion of active media in disordered structures may lead to stimulated emission of multiple scattered waves and then to the largely studied and debated random lasers. 5,6 Interesting developments of disordered photonics also include quantum effects such as the propagation of squeezed light in disordered systems or bunching of light in localized states. 19,20 The concept of spatially localized modes in a strongly disordered photonic structure can be interpreted in analogy with modes in an optical cavity. In both cases, standing electromagnetic waves are realized; although in disordered systems, the subtle balance that leads to a given photonic mode may strongly depend on the local dielectric environment. This idea has recently triggered the exploitation of disordered systems for reproducing experiments in nanocavity quantum electrodynamics (QED) by localization of light. 2,21 In this context, the possibility of introducing some degree of control of a random mode can be of the utmost relevance, due to the constraint on the spectral and spatial matching between emitter and photonic mode for maximizing QED effects. It has been theoretically shown that isolated random modes could be selectively tuned and possibly coupled to each other by a local variation of the dielectric structure. 22–24 The recent realization of artificial coupled states in a disordered system by nano-oxidation represents, therefore, a remarkable achievement in controlling random modes. 25 However, the spatial steadiness of a single random mode size, shape, and position as a function of the spectral tuning is still an open question. The relevance of this issue can be unfold, in a simplified picture, by the parallelism between disordered photonic media and photonic crystal nanocavities. In the latter, the existence of mirrors (bandgaps outside the lattice defect) assures that the localized modes are spatially confined within the cavity (lattice defect), even if a large perturbation of the system is performed for tuning the mode wavelength. 26–30 On the contrary, in disordered photonic media, the lack of mirrors and cavity implies that the mode spatial steadiness is not a priori guaranteed, whenever modifying the dielectric environment. In other words, the lack of photonic bandgap in disordered dielectric systems means that a localized random mode is usually embedded in a continuum of localized and delocalized modes. Therefore, the induced spectral tuning may produce hybridization with the surrounding modes.

In this letter, we address the experimental issue concerning the spectral tuning to a target wavelength of an isolated photonic mode localized in a strongly scattering disordered medium. In particular, we answer to the question whether the spatial modification of the disordered dielectric environment leads to the modification or even the destruction of the randomly shaped localized mode. We investigate a two-dimensional disordered photonic structure, where the randomness in position of the scattering centers is solely quenched by their finite size. This system has been previously studied demonstrating to support Anderson localized modes in the telecom wavelength range, which is relevant for applications driven by photonic research. 7,25 Near-field mapping of the emission of embedded quantum dots (QDs) is used to check the spatial distribution of the electric field. 25 Here, we demonstrate that an individual random mode can be finely blue shifted to a desired target wavelength by local photo induced oxidation, without altering its spatial distribution. The presented tuning technique is a proof of principle for any other gentle method based on local modification of the refractive index. Therefore, our findings prove the spatial steadiness of localized modes in a disordered photonic system.

The sample under study is fabricated on a 320 nm thick GaAs square suspended membrane of 25 µm x 25 µm size. Electron beam lithography and reactive ion etching are used to pattern the planar waveguide with nominally identical air holes (diameter 220 nm) giving an overall filling fraction of 30%. Finally, wet etching of an AlGaAs sacrificial layer is used to release the membrane. 31 We designed the position of the holes by a random sequential addition generator, 32 but we imposed a minimum distance (1.3 hole diameters) between the centers of the nearest neighboring holes, in order to avoid merging between adjacent holes during the fabrication process. Therefore, even though the sample is not fully random, hereafter, we will use the denomination random system/random modes both to stress the maximum practical randomness of our realization and to distinguish our sample from disordered media with higher spatial correlation used in different
context of light localization.\textsuperscript{2,3,17,33} The planar waveguide includes three layers of high-density (10\textsuperscript{3} \(\mu\text{m}^{-2}\)) InAs QDs, grown by molecular beam epitaxy, which are embedded in the middle plane of the slab. The experimental apparatus is based on a commercial scanning near-field optical microscope (SNOM), (TwinSNOM, Omicron NanoTechnology GmbH, Germany). The QDs are excited through a dielectric near field tip with a 780 nm diode laser at a power equal to 60 \(\mu\text{W}\). The photoluminescence (PL) signal is collected through the same tip, dispersed by a spectrometer, and detected by an InGaAs array. The high QDs density guarantees a homogeneous emission in a broad spectral range, from 1.24 \(\mu\text{m}\) to 1.34 \(\mu\text{m}\), all over the sample. Then, the observation of bright spots by measuring the map of the QDs PL can be directly related to the presence of a localized mode, which increases the QDs radiative rate. This is a relevant point of our experiment: the PL intensity collected by the tip is proportional to the electric field intensity of the photonic mode at the tip position, weighted by the spatial resolution of the system. Therefore, we obtain hyperspectral imaging of the photonic random modes with a combined spatial and spectral resolution of 250 nm and 0.1 nm for the optical signal, respectively. In addition to the spectral and spatial distributions of the optical modes, SNOM experiments also provide morphological information on the sample surface (with a spatial resolution better than 50 nm) via the mechanical feedback signal of the shear-force probe, which allows a precise alignment of subsequent maps. In order to control the disordered dielectric environment, we exploit a post-fabrication technique based on laser-assisted nano-oxidation that permanently and locally modifies the dielectric slab by concurrently reducing the effective GaAs membrane thickness and increasing the effective pore diameter.\textsuperscript{30}

In Fig. 1(a), we report scanning electron microscope images of the investigated sample. The inset displays the sample cross section and shows the suspended GaAs membrane with the patterned region. In order to avoid regions with higher optical losses and lower probability of mode localization, during the SNOM scans, we exclude the areas close to the borders of the lithographic pattern and we investigate inner sectors only. The PL intensity maps evaluated at fixed wavelength show several spots in different positions that arise from different localized modes. In order to address the spatial distribution of individual modes, we fully exploited the hyperspectral PL mapping by using different colors for different spectral resonances and largely separated modes. The resulting distributions are reported in Figure 1(a) for \(\lambda = 1293\) nm, in different colors for different modes. In Fig. 1(b), we report the PL spectra of the four more intense PL spots in the map at \(\lambda = 1293\) nm.

![FIG. 1. (a) Top view SEM image of the investigated GaAs slab of random air holes. The cross section of the slab waveguide is reported in the inset. The top view is reported with a superimposed SNOM PL optical map evaluated at 1293 nm, where different colors are used for different spectral resonances and largely separated modes. The scale bars are 1 \(\mu\text{m}\). (b) Near-field PL spectra normalized for the average PL of the whole scan and collected in center of the four different modes highlighted with the corresponding colors in the map in (a).]
FIG. 2. (a) Schematics of the laser assisted oxidation GaAs slab through the SNOM probe. ((b) and (c)) Near-field PL spectra of the red and green mode of Fig. 1 before and after the oxidation process, respectively. For the tuned modes in (b) are also reported, as black curves, the Lorentzian line shape fit of the spectra. (d) PL blue-shift of six different modes localized inside the same 5×5 μm² area, after exposition time of 200 min, as a function of the distance from the oxidation spot. (e) Red mode wavelength shift as a function of the oxidize laser exposure time. The data are reported along with slab oxidation schematics. The oxide islands are much thinner than the membrane thickness and the draws are not in scale. By using lines with the same scale color of the maps of Fig. 1(a). For a better comparison, the PL spectra are normalized to the PL average value over the whole map. We note that the spectral distributions of random modes cover a wide range of wavelength, with Q factor ranging from 200 to 400. The resonances can be spectrally well isolated and the modes are strongly spatially localized. These features and the statistical analysis (not reported here, see Refs. 7 and 25) allow us to conclude that we deal with Anderson localization, although this property is marginal in this contest.

Following the laser-assisted non-thermal oxidation method developed in Ref. 30, we used the near-field probe to locally modify the dielectric environment of the random system under consideration, as schematically reported in Fig. 2(a). This method allowed us for smoothly tuning the peak wavelength of any given localized photonic mode. Therefore, we placed the SNOM probe on the spot corresponding to the maximum PL intensity at λ = 1293 nm (red spot in Fig. 1(a)) and we employed the 780 nm laser with a power of 1.45 mW. The catalytic effect of electron-hole pairs photo-generated at the surface of the slab waveguide produces a controlled laser-assisted oxidation of the GaAs membrane. Due to the larger molar volume of the oxide with respect to the GaAs, the illumination produces an oxide island on the sample, which is formed by consuming the GaAs layer, and it likely extends both above and below the GaAs-air surface and in the pores. Using a weak laser excitation gives us a nanometric control over the oxide layers, thus, resulting in a very gentle tuning of the random modes. Concerning photonic effects, since there is a large mismatch between the refractive index of the oxide and the one of the GaAs, the nano-oxidation results in a reduction of the effective optical thickness of the membrane and an increase of the effective pores diameter size. In photonic crystal cavities, this has been used to perform a post growth blue-tuning of the resonant modes. Figures 2(b) and 2(c) show the initial and final spectrum of the investigated random modes (red and green spots in Fig. 1(a)), respectively. The two modes are separated by 2 μm and are differently tuned. This comparison proves that the oxidation process is local and that the tuning does not change the spectral line shape of the modes. Figure 2(e) reports the position of the central wavelength of the red mode as a function of the laser exposure time. We chose a very small tuning rate (of the order of 10⁻⁴ nm/sec) to highlight the accuracy of the method. The data show that the resonance of a given random mode can be gently blue-shifted to a desired target wavelength, and in addition, the low thermal budget of the oxidation avoids large damage of the quantum dots. Note that, after the final oxidation, the peak wavelength is shifted by 1.2 nm, while the PL intensity is...
reduced only by a factor of 4, likely due to the presence of the oxide layer disturbing the near field collection/illumination. Eventually, we investigated the tuning of the random photonic modes as a function of the distance from the exposure spot. In particular, we analyzed localized modes whose resonant wavelength is also distant from 1293 nm and therefore do not appear in the map of Fig. 1(a). In Fig. 2(d), we report the total shift after 200 min for six different modes with distance ranging from 0 to 4.5 µm from the exposure point. Clearly, the oxidation decays as a function of the laser spot position, although the effect extends over few microns, likely due to charges diffusion in the GaAs membrane.

The intricate nature of light localization in disordered systems with respect to ordered nanocavities raises the question whether a local perturbation of the disorder dielectric environment results in a modification of the shape, size, or position of the localized modes. In order to answer this question, we perform near-field imaging of random modes as a function of the photo-induced oxidation time. In disorder based systems, there is a large spectral and spatial overlap of modes that can spoil the PL intensity map evaluated at a given wavelength, by introducing spots associated to broad tails of resonances that, unfortunately, are peaked at neighboring wavelengths. To overcome this issue, we performed a Lorentzian line shape fit of the PL emission. Figures 3(a) and 3(b) show the PL intensity maps and the amplitude of the Lorentzian line shape of the red mode at initial

![Fig. 3](image-url)
(t = 0 min), intermediate (t = 20 min), and final (t = 200 min) tuning, respectively. By exploiting the morphological information of SNOM measurements, we superimposed the optical mode maps to the corresponding SEM images, in order to precisely locate the disorder localized mode. The alignment of the SEM images to the PL maps is with an uncertainty of the order of 50 nm (the spatial pixel in the high resolution scans is 100 nm). It turns out that the three PL intensity maps are very similar. For a more quantitative analysis, the horizontal and vertical normalized cuts are reported in Fig. 3(d) for t = 0 and t = 200 min (the data at t = 20 min overlap almost perfectly with t = 0 and are not reported for clarity). The data show that the horizontal cuts are shifted by 20 nm and the vertical cuts by 50 nm (both values are within our uncertainty in the alignment). Therefore, the randomly originated mode does not significantly change its spatial distribution after the tuning process induced by local laser oxidation.

In conclusion, we have experimentally shown that controlling the wavelength of random photonic modes without altering the mode intensity distributions is possible. Exploiting GaAs oxidation by near-field laser exposure, we can blue-tune the resonance to a specific target wavelength. This is of the utmost relevance for specific application of disordered photonics, as, for example, cavity quantum electrodynamics experiments, where the constraint of spectral matching between emitter and photonic mode is mandatory. Demanding for these applications is also the robustness of the disorder localization to external perturbation, which we demonstrated by performing a gentle modification of the dielectric environment. Post-fabrication local control of the disordered photonic media with a low thermal budget can be therefore exploited to adjust the coupling strength between emitters and random photonic modes. Our results open the way to exploit in random systems other tuning methods (as liquid nanoinfiltration, anodic oxidation, dielectric deposition, etc) developed in the field of photonic crystal cavities, where also red shift and/or reversible approaches can be envisaged.

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