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Citation for published version (APA):

DOI:
10.1063/1.4712410

Document status and date:
Published: 01/01/2012

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

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Download date: 26. Dec. 2019
Transfer printing and nanomanipulating luminescent photonic crystal membrane nanocavities

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(Received 30 November 2011; accepted 9 April 2012; published online 4 May 2012)

The release of photoluminescent InGaAsP photonic crystal nanocavity chiplets from the host chip for creating autonomous functional microparticles is demonstrated. A transfer printing method using a soft polymeric material as a stamp is used to transfer cavity arrays to other substrates. Alternatively, cavities are transferred individually by a nanomanipulation technique. The chiplets can be fully deterministically positioned on both the host chip and another substrate (glass) with the nanomanipulator. The chiplets have the striking property of spontaneously orienting themselves with their plane perpendicular to the receiving surface. At each stage of the process, the condition of the cavities as dependent on their immediate surroundings is monitored from their photoluminescence spectrum. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4712410]

I. INTRODUCTION

In recent years, there has been an increasing interest in releasing opto-electronic circuits, made by state of the art semiconductor technologies in wafer-scale crystalline host materials, from their substrates. New applications are enabled when the dedicated structures can be fabricated on an arbitrary substrate or even can function as free particles. One technique is based on transfer printing using an intermediate soft polymer as a stamp; a technique which may be used for heterogeneous integration of very thin crystalline layers of dissimilar semiconductors.1 If the functional unit is small enough, it can function on a diversity of flexible or curved surfaces,2 which may include the human skin.3 With a multilayered structure, deposited on a prepatterned stamp, transfer printing has recently been used to create novel three-dimensional optical metamaterials.4 A nanobeam type photonic crystal (PhC) nanocavity was shown to still operate as a laser after being transferred to a flexible substrate.5 The individual micromanipulation and aligned stacking of small-area nanopatterned structures has yielded three-dimensional (3D) PhCs,6 which recently led to the demonstration of the coupling of a quantum dot (QD) to a 3D PhC cavity7 and subsequently to the first 3D PhC nanolaser.8 An individual planar PhC nanocavity has been transferred from the original wafer and subsequently employed as a micromanipulated device for coupling to QDs that reside on a foreign substrate.9 Recently, the site-specific transfer-printing of individual graphene microscale patterns to arbitrary surfaces by a micro-manipulator has been demonstrated.10 Layered structures, released as particles from the chip by brute force methods such as sonication, have served as sensors in suspension-based multiplexed bio-assays.11 Because the latter particle structure behaves as an optical cavity with a spectrum that serves as an identification code and with which the refractive index of its environment can be sensed, these particles are referred to as “smart dust.”11

In the present work, we report planar PhC nanocavities made in a thin semiconductor membrane and suspended by breakable tethers so that they can be easily released after fabrication as chiplets. Luminescent QDs are embedded in the semiconductor membranes which serve as internal light sources to excite the cavity modes. The cavity spectrum can thus be read out remotely from the stand-alone cavities by detecting the photoluminescence (PL). Both transfer printing and deterministic positioning by individual nanomanipulation are demonstrated. A remarkable preference for the perpendicular orientation of the cavities’ plane and the receiving surface is observed.

QDs and other nanoparticles, made by bottom-up approaches, have by now an established position in a wide variety of applications, ranging from medical diagnostics, medical treatment, and medicine administering,12 to solar energy harvesting.13 On the other hand, using the top-down processes from the semiconductor industries, dense electronic or optical integrated circuits are manufactured on a wafer scale with dimensions approaching nanometer scale definitions and precision. The entities reported in the present work are first manufactured using nanolithographic processes on a chip and are then released from the host chip. The microscopic environment of the particles, including their bonding to the substrate, determines the cavities’ resonance spectra, which can be exploited in applications. Thus a microns sized chiplet is obtained that serves as an autonomous particle with intelligent functions. The functionality of, and the communication with, the particle is provided by the nanophotonic circuits imprinted on it.
There exists strong interest in combining the unique optical properties of expensive, layered epitaxially grown and surface patterned III–V crystalline semiconductors with more common and cheap materials as glass, plastic, or silicon. The key advantages of the presented techniques are that active, light-emitting particles can be easily integrated with optical circuits based on such cheap materials. Their smallness allows them to be incorporated in flexible circuits on polymer substrates with bending radii very much larger than the particles’ size of a few microns. Only minute amounts of the dedicated material are needed. When only one or very few particles are needed on a given substrate, the nanomanipulation technique is most suitable, as it also allows easy alignment with structures on the substrate. As one simple application, the attachment of such a particle to an optical fiber tip for sensing applications has been demonstrated. When patterns of chiplets are needed, the printing technique would be preferable, with possible further processing after transfer to the receiving substrate. The chiplets also could function completely autonomously in a similar way as the multilayer structures of Ref. 11, as encoded particles in suspension-based bio-assays. For the building of three-dimensional structures from these membranes, the vertical orientation could be exploited as a new building tool.

II. FABRICATION AND MEASUREMENT

PhC nanocavities were fabricated in a 220 nm thick InGaAsP membrane which contains a single layer of self-assembled InAs QDs (density $3 \times 10^{10}$ cm$^{-2}$). The pattern was defined by high resolution electron beam lithography on a 350 nm thick ZEP 520 A resist and subsequently transferred to an underlying 400 nm thick SiN$_x$ mask layer by reactive ion etching. Next, the pattern was etched in an InP-InGaAsP-InP layer stack, by inductively coupled plasma etching using Cl$_2$/Ar/H$_2$ chemistry. A free standing InGaAsP membrane was achieved by selective wet chemical etching of InP using an HCl:H$_2$O = 4:1 solution at 2 °C. H$_0$-type cavities are made, defined by modifying four neighboring holes without leaving any holes out, see the scanning electron microscope (SEM) image of a fabricated nanocavity in Figure 1(a). To enable the removal of the nanocavities from the parent chip, every nanocavity was weakly suspended by two tether bars.

These cavities are chosen because they belong to the smallest possible PhC cavities, are well characterized, and have reasonably high cavity $Q$-factors. They were used previously in our lab to demonstrate the versatility of spectral encoding. The PhCs have a lattice spacing (a) of 500 nm and a nominal hole diameter of 320 nm (radius-to-lattice-spacing ratio r/a = 0.32). This leads to a calculated photonic bandgap between approximately 1400 and 1700 nm wavelength. The QD peak emission was at 1550 nm, but due to size dispersion and temperature broadening, the total emission bandwidth spans the photonic bandgap so that we have a broadband source that can excite any cavity resonance in the bandgap. The H0 cavity has a number of modes (typically four) of which the frequencies can be varied across the bandgap, depending on the parameters of the modified holes. For the present purpose, the choice of the resonant frequencies was somewhat arbitrary, and no optimization of $Q$-factors was attempted. The diameters of the modified nearest four neighbor holes are 230 nm. Two of them are shifted along the x direction by 70 nm (0.14 a) and the other two by 20 nm (0.04 a) in the y direction. Since the cavity mode’s evanescent field extends inside the holes and up to some 100 nm outside the plane surface, the resonant frequencies also depend on the presence of dielectrics on the cavity surface or inside the holes. Frequency shifts after introducing dielectrics of a given refractive index at different positions in the cavity were calculated using three-dimensional finite difference time domain simulations.

Comparison of these calculated shifts with the measured ones allows conclusions on the close proximity and area coverage of the receiving substrate surface to the cavity surface or on the infiltration of the airholes.

PL measurements were performed using a continuous wave excitation laser at a wavelength of 660 nm. An optical microscope objective with NA = 0.5 and magnification of 50 is used for both excitation of the cavities and collection of the PL. After dispersing the PL in a 50 cm focal length monochromator, the collected signal is detected by a liquid nitrogen cooled InGaAs array, which has a long wavelength cut off at 1600 nm.

III. RELEASING METHODS

A. The transfer printing method

As the first experiment, the cavities are handled by a transfer printing process. Several soft materials were tried as stamps, including soft polymer sheets or adhesive tape, which all worked similarly well. The foil was gently pressed to the host substrate, and after being pulled back, the cavities stuck to the foil. While the cavities were on the foil, the PL spectra were measured, which shows the functionality on a flexible substrate and is of interest per se. Subsequently, the foil was transferred to the receiving substrate, typically glass. The results are shown in Figure 2, where an adhesive tape was used as a stamp.

Figure 2(a) shows the original array before release. Figure 2(b) shows the same array pressed on the glass substrate but with the tape not yet removed. A gentle pressure was exerted to press the cavities to the glass. Figure 2(c) shows the same array after removing the tape. Only a subset
of eight cavities adhered to the glass, the rest remained on the tape. It was found that the bonding strength to the glass is strongly dependent on the exerted pressure, which was non-uniform. For the present work, no effort was made to optimize the printing process in order to maximize the yield. Apparently, for the small area devices used, no adhesive is necessary, which is different from the heterogeneous integration process that employs large area die to die bonding.21 In Figure 2(d), the PL-spectra of one of the cavities are shown under the three conditions of Figures 2(a)–2(c). The original spectrum, shown in the lower trace in Figure 2(d), can be well identified by the simulations, but this will not be discussed here. All peaks are strongly redshifted for the situation in Figure 2(b); see the top trace of Figure 2(d), taken from the transparent glass side. From simulations, it follows that all holes as well as the top and bottom environments around the cavity are filled with a dielectric of typical refractive index around 1.5. This means that the glue from the tape has been pressed into the holes. When the tape is removed, the spectrum shifts back closer to its original value, see Figure 2(d), the middle trace. The remaining redshift can be explained by the presence of the glass on one side of the cavity. These observations prove that the cavities make intimate contact with the glass, and the bond is strong enough to withstand the release of the tape and associated removal of the glue from the holes. It was found that the cavities indeed are rather robust against mechanical perturbations. It is expected that the bond, which in absence of other forces will ultimately be determined by Van der Waals (vdW) forces, is so strong because the thin membranes are somewhat flexible and, therefore, conformally follow the glass surface.

B. Nanomanipulation

In the next experiment, individual PhC nanocavities were transferred and manipulated. A tapered glass fiber with a tip diameter ~500 nm (see the inset of Figure 1(b)) is actuated by a three-axis piezo-controlled nano-manipulator. Figure 1(b) shows the scheme of the nano-manipulation set-up. With the tip, a gentle force is exerted on the nanocavities so that the tethers are broken. Then the nanocavities are released from the chip as chiplets. They attached spontaneously to the tip without any additional effort. The orientation of the cavity plane with respect to the taper shaft was found to be random as observed under the microscope. The position and orientation of the chiplet on the tip can also be noted from the spectral shifts in the cavity spectra as shown in Figure 3. The shifts vary from smaller than the spectrometer resolution (wavelength shift <0.1 nm) when the tip is near the edge to ~10 nm when the tip is attached to the center of the surface of the cavity. With the tip near the edge, it is well outside the intensity distribution of the cavity mode and so does not affect the resonance frequency. With the tip near the center and pressed to the chiplet, it is inside the evanescent field of the mode, so that this extra refractive index will redshift the resonance.

The chiplets were subsequently transferred from the taper tip to a glass plate with their face parallel to the glass surface. With the cavities just being placed on the receiving substrate, they usually could still be moved over the surface by pushing them with the tip. After pushing them gently onto the surface using the fiber tip, they firmly attach to the glass. Once they stuck well, they could not be moved again with the tip, as is the case for the printed chiplets in Figure 2.

The PL spectrum in this case shows a redshift compared to the original situation in the parent chip, similar to the case in Figures 2(c) and 2(d).

FIG. 2. (a) A microscope image of an array of nanocavities on the host chip. (b) The same array, after removal by the tape and pressed to the glass substrate. The image was taken from the glass side, so it looks as a mirror image of (a). The tape is not yet removed. (c) The same array, after the tape is removed, again seen from top, so same view as in (a). Only 8 cavities are transferred, the rest remained on the tape. Note some damage to one of the cavities in (b) and (c), indicated by the arrows, that may serve as additional recognition mark. (d) Spectra from one of the cavities (encircled) in situation (a) lowest trace, (b) upper trace, and (c) middle trace.

FIG. 3. Microscope images from of (a) a chiplet attached to the tip with its edge and (b) a tip attached to the central part of the chiplet. The insets of (a) and (b) show sketches of the two cases. (c) and (d) show the corresponding PL spectra (green upper curves) of the two cavities shown in (a) and (b). The PL spectra with the chiplets suspended on the parent chip (red lower curves) are also included for comparison.
C. Discussion

Both the transfer printing and the fiber manipulation technique depend on a balance of the forces between the parent substrate, the fiber manipulator, and the receiving substrate. After the tethers are broken, the forces between the different bodies are determined by both the nature of the force as well as by the area over which the two bodies are in contact. The contact forces can be of electrostatic nature, originate from capillary forces due to an adsorbed water film on the surface from the atmospheric environment, or ultimately result from the Van der Waals forces. Below, it is shown that even the weak Van der Waals forces are enough to explain the strong bonds of the particles to surfaces. The nature of the forces cannot be changed, but the contact areas can.

For the printing process, the bond to the “stamp” must be strong enough to break the tethers. Then the bond to the receiving substrate must be stronger than to the stamp. This is a delicate and nontrivial process, which is not known in detail and was not optimized. It is assumed that the contact area with the microscopically rough stamp material plays a crucial role here. The delicate balance is assumed to be responsible for the relatively low yield of transfer in the current stage, in the order of 30%, as follows from a comparison of Figs. 3(a) and 3(b).

In the case of the fiber manipulation, there is more control on the contact area. In practice, cavities were taken out by a two-step process. In the first step, the cavities were punched out by breaking the tethers, then fell into the undercut void, no longer in contact with the fiber tip. Since the void has a non-flat shape, the membrane only made contact at a few spots. Approaching again with the fiber tip, the contact area with the fiber tip was adjusted, until the cavities attached to the fiber. Releasing the cavity on the receiving substrate was a trial-and-error process of adjusting the fiber: moving it across the cavity surface while it already is in contact with the receiving substrate, either flat or upright. Again, this process is not known on a microscopic level but works fairly smoothly in practice.

IV. SELF-ORIENTATION ON SURFACES

Striking and unexpected phenomena were observed during the manipulation. Accidentally, chiplets jumped from their positions and landed on the surface some tens of microns from their original positions. Rather than landing in the intuitively expected orientation with the face parallel to the substrate. After the tethers are broken, the forces between the different bodies are determined by both the nature of the force as well as by the area over which the two bodies are in contact. The contact forces can be of electrostatic nature, originate from capillary forces due to an adsorbed water film on the surface from the atmospheric environment, or ultimately result from the Van der Waals forces. Below, it is shown that even the weak Van der Waals forces are enough to explain the strong bonds of the particles to surfaces. The nature of the forces cannot be changed, but the contact areas can.

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for the genuine vertical orientation, as found from observations looking perpendicularly to the surface.

By careful handling of the taper tip, balancing the interaction of the chiplet with the tip and substrate surface, the vertically standing chiplets can be fully maneuvered. The chiplets can be dragged by the tip to different positions, after which the tip can be removed. It is possible to lift the standing chiplets from the substrate with the tip and then place them back on the substrate on a predetermined position. Also, by touching the standing chiplet away from its center, it can be rotated along an axis in its plane, perpendicular to the substrate surface, with one edge always in contact with the substrate. With all these controls, a fully deterministic positioning is possible. This was demonstrated by lining up a number of vertically standing chiplets, all with their surfaces oriented parallel, on the semiconductor surface as in Figure 4(c) or on a glass plate as shown in Figure 4(d). It is emphasized that the manipulations were performed under a large working distance and a low-resolution optical microscope. SEM was only used for taking images not for manipulation. No efforts were done to optimize the process or make devices based on a single or ensembles of vertically positioned chiplets.

It is expected that the counter-intuitive observations of the robustness of the particles’ bonding to the surface in any orientation are due to the small areas involved. On that scale, the surfaces are relatively smooth and therefore the adhesion forces are strong. Ultimately, the force between two surfaces is determined by the Hamaker constant, which typically is on the order of $10^{-19}$ N, and $S$ is the contacting area. This force should be compared to the particles’ weight, which can be estimated as $W \sim 10^{-12}$ N. The smallest possible area relevant for $S$, applicable for the situation of Figure 4(b), is the cross-sectional area of the tether, $\sim 200 \times 200$ nm$^2$. With surfaces in contact, which means $d \sim 0.2$ nm, the vdW force on this small area is in the order of $2 \times 10^{-5}$ N or $10^7$ times its weight. Alternatively, when
the tether tip is near 50 nm from the surface, the attractive force to the surface equals the particles’ weight. The attractive force between edge or tether tip can provide the torque to rotate the particle perpendicular to the surface, when it slowly approaches the surface as when attached to the fiber. Another force could come from static charging of the particle after release, either through its interaction with the glass fiber or from charge separation in the semiconductor before the release. Since the charge will be concentrated near the extremities of the sample, the interactions with its image charges in the substrate will exert a torque that rotates the chiplet in the vertical position. This mechanism could be particularly relevant for the throwing experiments through air, since the long range of the electrostatic force will enable the rotation of the particle during its flight. After landing, however, no indications for static charging were found.

V. CONCLUSION

In conclusion, we have introduced both the transfer printing method and a nano-manipulation technique to release InGaAsP photonic crystal nanocavity chiplets from their parent chip. Since the cavities are luminescent, they can be read out remotely in free space using either objectives or fiber-optic components. They can, thus, be employed as autonomous particles that perform a variety of useful functions when transferred to a foreign substrate. Examples include the function as sensors, for example, refractive index or temperature, either when attached to a foreign surface or from charge separation in the semiconductor before the release. Since the charge will be concentrated near the extremities of the sample, the interactions with its image charges in the substrate will exert a torque that rotates the chiplet in the vertical position. This mechanism could be particularly relevant for the throwing experiments through air, since the long range of the electrostatic force will enable the rotation of the particle during its flight. After landing, however, no indications for static charging were found.

ACKNOWLEDGMENTS

The authors acknowledge E. J. Geluk, P. J. van Veldhoven, B. Smalbrugge, P. Nouwens, and T. de Vries for their help in the fabrication processes. We thank A. Fiore and E. P. A. M. Bakkers for useful discussions. The support from the BrainBridge project (Zhejiang University-University of Eindhoven University and Philips Research collaboration) is gratefully acknowledged.