Printed Assemblies of Inorganic Light-Emitting Diodes for Deformable and Semitransparent Displays

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We have developed methods for creating microscale inorganic light-emitting diodes (LEDs) and for assembling and interconnecting them into unusual display and lighting systems. The LEDs use specialized epitaxial semiconductor layers that allow delineation and release of large collections of ultrathin devices. Diverse shapes are possible, with dimensions from micrometers to millimeters, in either flat or “wavy” configurations. Printing-based assembly methods can deposit these devices on substrates of glass, plastic, or rubber, in arbitrary spatial layouts and over areas that can be much larger than those of the growth wafer. The thin geometries of these LEDs enable them to be interconnected by conventional planar processing techniques. Displays, lighting elements, and related systems formed in this manner can offer interesting mechanical and optical properties.

Display devices represent ubiquitous, central components of nearly all consumer electronics technologies. Organic light-emitting diodes (OLEDs) are rapidly emerging as an attractive alternative to backlit liquid crystals due to their comparatively high refresh rates, contrast ratios, power efficiencies, and capacity for vibrant color rendering (1, 2). Inorganic light-emitting diodes (ILEDs) can also form displays, with properties such as brightness, lifetime, and efficiency that can exceed those possible with OLEDs (3, 4). These displays exist, however, only in ultralarge-area, low-resolution formats (square meters; billboard displays), limited by processing and assembly procedures that do not scale effectively to small (<~200 μm by 200 μm), thin (<~200 μm) light emitters or to dense, high-pixel count arrays. An ability to replace existing

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**Fig. 1.** (A) SEM image of a square array of AlInGaP LED structures (50 μm by 50 μm) created by vertical, patterned etching through an epitaxial multilayer stack grown on a GaAs wafer. (B) Cross-sectional SEM view of one of these structures, showing the LED semiconductor layers (quantum wells, as well as cladding, spreading, and contact layers) on a sacrificial epilayer of AlAs. (C) Schematic illustration of a printing-based assembly method for transferring collections of LEDs (gray) released from the GaAs wafer to a target substrate (shown here as a flexible sheet). (D) SEM image of the GaAs wafer after removing a set of LEDs (indicated by white arrows) with a stamp. (E) SEM image of a region of the target substrate printed with this stamp. (F) Angled-view SEM image of an individual LED (i.e., ILED) from the array in (D). A pair of “breakaway” photoresist (PR) anchors at the two far corners of the device holds it above the GaAs wafer in the suspended configuration of a diving board, for ease of liftoff with a stamp. The white arrow points to the region of removed AlAs. (G) SEM image of a dense collection of such devices on a piece of a GaAs wafer. The black arrow and white dot indicate, roughly, the region of this chip that corresponds to the image of (F). (H) Optical image of a target substrate printed with sparse arrays of devices at different spacings, derived from the chip shown in (G). (I) Large-scale collection of ILEDs (1600 devices, in a square array with pitch of 1.4 mm) printed onto a thin, flexible sheet of plastic, shown here wrapped onto a cylindrical glass substrate (main panel). The inset shows a similar collection of ILEDs (1600 devices, in a square array with pitch of 1.4 mm) printed onto a plate of glass. For these cases, relatively large ILEDs were selected for ease of viewing; devices with dimensions of (E) are invisible at this magnification.

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methods for fabricating ILEDs (i.e., wafer sawing, serial pick-and-place, wire bonding, and packaging on a device-by-device basis) and for incorporating them into displays (i.e., robotic assembly into tiles followed by interconnection using large quantities of bulk wiring) with those that more closely resemble the planar, batch processing of OLEDs would greatly expand the application opportunities. Examples include not only ILED displays for desktop monitors, home theater systems, and instrumentation gauging, but also, when implemented in flexible or stretchable forms, wearable health monitors or diagnostic and biomedical imaging devices. In microscale sizes, such ILEDs can also yield semitransparent displays, with the potential for bidirectional emission characteristics, for vehicle navigation, heads-up displays, and related uses. We present routes to create ultrathin, ultra-small ILEDs in flat or "wavy" geometries and to assemble them into addressable arrays using scalable processing techniques, on substrates ranging from glass to plastic and rubber. The strategy involves four key components: (i) epitaxial semiconductor multilayers designed for lateral delineation and release from a source wafer to yield isolated arrays of ILEDs, each of which remains tethered to the wafer by polymeric "breakaway" anchor structures; (ii) printing techniques for manipulating the resulting ILEDs in schemes that enable formation of large-scale arrays on foreign substrates and in arbitrary spatial layouts; (iii) planar processing methods for establishing electrical interconnects to the devices, in direct or matrix addressable configurations; and (iv) integration strategies capable of yielding ILED displays in flexible or stretchable formats and with conventional, semitransparent, or bidirectional emission characteristics. Certain aspects build on previously reported procedures for etching and manipulating epitaxial semiconductor layers (5–11) and for fabricating flexible and stretchable electronics (12, 13).

Figure 1 presents essential aspects of the first two of the components [see supporting online material (SOM) for details]. The epitaxial semiconductor layers include AlInGaP quantum well structures (6-nm-thick In0.53Ga0.47P wells, with 6-nm-thick barriers of Al0.25Ga0.75In0.25P on top and bottom), cladding films (200-nm-thick layers of In0.75Ga0.25P:Zn and In0.5Al0.5P:Si for the p and n sides, respectively), spreaders (800-nm-thick layers of Al0.45Ga0.55As:C and Al0.45Ga0.55As:Si for the p and n sides, respectively), and contacts (5-nm-thick layer of GaAs:Si for the p and n sides, respectively), for a total thickness of ~2.523 μm, all grown on AlAs (1500-nm-thick layer of Al0.09Ga0.91As:Si) on a GaAs substrate (fig. S1). The AlAs can be removed by etching with hydrofluoric (HF) acid, in procedures that do not alter the overlying layers or the underlying substrate. The process for defining the ILEDs first involves forming a pattern of vertical trenches through the epitaxial layers by inductively coupled plasma reactive ion etching through a mask of SiO2 defined photolithographically (fig. S2). This step determines the lateral geometries of the devices (fig. S2). Figure 1A and B, shows top and cross-sectional scanning electron microscope (SEM) images collected after this etching process for a representative case, where the device islands in Fig. 1 are 50 μm by 50 μm. Creating a pattern of photore sist posts (i.e., "breakaway" anchors) located at two of the four corners of each island, followed by immersion in concentrated HF, leads to the undercut release of an organized array of ILEDs. The anchors hold the devices in their lithographically defined locations to prevent lift-off into the etching bath, even after complete undercut (fig. S2). Next, an automated printing tool (fig. S3) brings a soft elastomeric stamp with features of relief embossed onto its surface into aligned contact with a selected set of these ILEDs. Peeling the stamp away fractures the photore sist anchors and leaves the devices adhered via Van der Waals interactions to the raised regions of relief. Figure 1C and D, shows schematic illustrations of the printing process and an SEM image of an array of anchored ILEDs on the source wafer after one cycle of printing (fig. S4). The white arrows in Fig. 1D highlight the collection of ILEDs removed by this process, corresponding to every third device along the two orthogonal axes of the square array. Figure 1E provides an SEM image of these devices printed onto a glass substrate. The engineering design of the breakaway anchors is such that they are sufficiently robust to hold the ILEDs in their representative device before undercut etching on the GaAs wafer, and after transfer printing onto a polyurethane-coated glass slide. The inset provides a histogram of the bias voltages needed to produce currents of 0.1 mA in a collection of devices. (E) Spectral characteristics of emission for a typical device on the wafer and after transfer printing.
Fig. 3. (A) Schematic illustration of a planar scheme for interconnecting a printed array of ILEDs in a passive matrix layout. Coordinated control of voltages applied to the row and column electrodes allows operation in a passive matrix display mode. (B) Images of a flexible display that incorporates a 16 by 16 array of ILEDs in the layout shown in (A), on a sheet of plastic (PET), wrapped around the thumb of a mannequin hand (main panel; human scale; radius ~8 mm) and a cylindrical glass tube (inset; radius ~12 mm). External interface to control electronics occurs through ribbon cables bonded to column and row electrodes that emerge from the periphery of the display. (C) Image of a comparatively large, semitransparent display that uses a similar layout but with a sparse array of ILEDs on a glass substrate. The camera is focused on the paper in the background; the white dashed box illustrates the perimeter of the active region of the display. (D) Image of a similar device (bottom right) displaying a different pattern in front of a mirror (upper left), to illustrate the bidirectional emission property. In this system, the ILEDs represent only ~0.8% of the total area. The inset shows a magnified view of a region of the display in its off state, to illustrate the small areal coverage of the devices. The black arrow points to one of the ILEDs, which is barely visible at this magnification.
Two spin-cast, photopatterned layers of epoxy (1.2 μm thick) provide openings to these contacts; the top layer electrically separates the column and row electrodes at their crossing points. Connecting terminal pads at the ends of these electrode lines to external computer control systems via ribbon cables that use anisotropic conductive films (ACFs) enables passive matrix addressing (see SOM and fig. S8 for details). Figure 3B shows images of a small display that uses this design, formed on a thin sheet of plastic (PET, 50 μm thick) with a layer of a photocurable polyurethane as an adhesive. The ILEDs have dimensions of 100 μm by 100 μm and are configured into a 16 by 16 square array. The yields on the individual pixels for the case of Fig. 3B are 100%; at the level of the display, one column and two rows do not function, due to breaks in the contacts to the ACF ribbon cable [fig. S9; see SOM and fig. S10 for an example of similar display with even smaller ILEDs (50 μm by 50 μm)].

Such systems can be bent to radii of curvature of ~7 mm, with no observable degradation, even for several hundred cycles of bending (fig. S11). Analytical calculation shows that even at the minimum bend radius investigated here, the maximum strain in the ILED is 0.21%, with a somewhat smaller strain (0.19%) in the quantum well region (see SOM for details). Analysis using literature parameters to determine the dependence of the bandgap on strain (19–22) suggests changes in emission wavelength of ~2.4 nm for the smallest bend radius (see SOM for details). As shown in Fig. 1, step-and-repeat printing can yield systems that cover areas much larger than those of the constituent ILEDs or the source wafer. One important outcome is the ability to form displays that can offer an effectively high level of transparency, where only the ILEDs (and the electrodes, if they are not made with transparent conductors) are opaque. Figure 3, C and D, shows examples of a 16 by 16 array, formed on glass. Here the area of the display is ~325 mm²; the cumulative area of all the ILEDs is only ~2.5 mm², corresponding to less than ~1% of the display area. Figure 3C illustrates the operation of such a system positioned above a sheet of paper with printed logos; the focus of the image is on the paper, thereby illustrating a practical level of transparency for application in a heads-up display, for example. Figure 3D shows the same device (lower right), operating in front of a mirror (upper left) to demonstrate bidirectional emission characteristics. The inset provides a magnified view of a region of this display, in its off state to show the small sizes of the ILEDs compared to the unit cells. These layouts are critically important for many applications, due to the efficient utilization of the LED material, for reduced cost. For the examples shown, we achieved ~98% yields on the individual devices, and ~80% yields on the interconnections, limited by breaks in the metal lines and failed contacts to the ACF ribbon cable (fig. S12).

The devices and integration methods reported here are compatible with strategies to produce stretchable electronics (12, 13), thereby providing a route to conformable displays and lighting systems of the type that might be interesting for integration with the human body and other curvilinear, deformable surfaces, all of which demand more than simple bending (e.g., Fig. 3B).

Figure 4A shows an example of a stretchable ILED with the shape of a ribbon. This device was formed by transfer printing and bonding to a prestrained, rubber substrate of PDMS. Relaxing the prestrain creates a device with a “wavy,” sinusoidal profile; this structure responds elastically to applied strain with a physics similar to that of an accordion bellows (12, 23) to yield a stretchable ILED device. The top panels provide finite element simulation of the mechanics of the system in compressed (left) and stretched (right) configurations. The results indicate maximum strains in the ILED and the quantum well region of 0.36 and 0.053%, respectively (see SOM for details). The bottom panels show optical micrographs in the off (top) and on (bottom) states, with and without external illumination, respectively, in configurations similar to those illustrated in fig. S18A. The emission characteristics show no noticeable change in color with applied strain or associated changes in device geometry from “wavy” to flat (see SOM and figs. S13 and S14 for details). This observation is consistent with a calculated change in emission wavelength of less than ~0.7 nm based on our computed strain values and analysis similar to that performed for the flexible display (see SOM for details).

![Voltage (V) needed to generate a current of 20 μA measured after stretching cycles to 500 times at an applied strain of 22%. The inset shows the I-V behavior after these cycling tests. These devices have relatively high turn-on voltages, due to the use of nonohmic contacts.](www.sciencemag.org)
The “wavy” strategy of Fig. 4A can accommodate only a relatively modest range of applied strains (i.e., up to a few percent, for the designs reported here). A path to displays with high levels of stretchability uses non-coplanar mesh designs adapted from schemes reported for integrated circuits (13). Figure 4B presents optical micrographs of such a system, composed of a 16 by 16 square array of ILEDs bonded to a PDMS substrate and interconnected by electrodes supported by arc-shaped bridges, with a fraction of the pixels turned on (overall yield >80%) (see SOM and fig. S15 for details). The shapes of these bridges change in response to deformations of the display, in a way that isolates the ILEDs from any significant strains (figs. S16 and S17). In particular, calculation shows that for strains of 24%, as defined by the change in separation between inner edges of adjacent device islands, the maximum strain in the ILED and quantum well are only 0.17 and 0.026%, respectively. The computed change in emission wavelength is less than ~0.3 nm (see SOM for details). Figure 4C provides optical micrographs of four pixels in this display, in their off and on states, with (top) and without (bottom) external illumination, respectively, in compressed and stretched configurations. The images show the expected reduction in the heights of the arc-shaped bridges that lie in the direction of the applied tensile force (i.e., along the interconnects that run from lower left to upper right), together with an increase in the heights of the bridges in the orthogonal direction, due to the Poisson effect. This mechanical response is fully elastic—the bending-induced strains in the interconnects are small, the strains in the ILEDs are negligible, and the strain in the PDMS is well within its linear response regime. The data in Fig. 4, D and E, are consistent with these mechanics, as are the associated mechanics calculations. In particular, the current-voltage characteristics of a typical device do not change in a measurable way for applied strains up to ~22%, and we observe no degradation on cycling up to a few hundred times (500 times). Recent work demonstrates the use of smaller collections of large, conventional ILEDs in deformable devices that use different designs (24, 25).

The schemes reported here for creating thin, small inorganic LEDs and for integrating them into display and lighting devices create design options that are unavailable with conventional procedures. The planar processing approaches for interconnect resemble those that are now used for organic devices and, for example, large-area electronics for liquid crystal displays, thereby conferring onto inorganic LED technologies many of the associated practical advantages. In large-area, high-pixel count systems (e.g., 1 million pixels per square meter), the ability to use LEDs with sizes much smaller than those of the individual pixels is critically important to achieve efficient utilization of the epilayer semiconductor material, for reasonable cost. The minimum sizes of devices reported here are limited only by the resolution and registration associated with manual tools for photolithography.

References and Notes

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Supporting Online Material

www.sciencemag.org/cgi/content/full/325/5943/977/DC1 Materials and Methods

Figs. S1 to S19

References

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Visualization of Fermi’s Golden Rule Through Imaging of Light Emission from Atomic Silver Chains

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Atomic-scale spatial imaging of one-dimensional chains of silver atoms allows Fermi’s golden rule, a fundamental principle governing optical transitions, to be visualized. We used a scanning tunneling microscope (STM) to assemble a silver atom chain on a nickel-aluminum alloy surface. Photon emission was induced with electrons from the tip of the STM. The emission was spatially resolved with subnanometer resolution by changing the tip position along the chain. The number and positions of the emission maxima in the photon images match those of the nodes in the differential conductance images of particle-in-a-box states. This surprising correlation between the emission maxima and nodes in the density of states is a manifestation of Fermi’s golden rule in real space for radiative transitions and provides an understanding of the mechanism of STM-induced light emission.

The scanning tunneling microscope STM, which is based on the tunneling effect, has been used to visualize various quantum phenomena in real space, including the quantum corral (1), quantum mirage (2), and particle-in-a-box states (3, 4). All of these demonstrations involved the localization of the electron density of states in confined nanostructures. Light emission from the STM junction reveals a different kind of quantum phenomenon that involves the optical transitions and inelastic electron tunneling (IET) processes in single molecules (5, 6) and nanostructures (7). Furthermore, photon intensity imaging with atomic resolution has been demonstrated (8–10). The spatial resolution in these optical experiments originates from the precision of the STM in injecting electrons in a confined space, although the emitted photons are collected in the far field. This atomic-scale optical detection can reveal aspects of the molecules and nanostructures that are hidden when probed with other techniques. Imaging of STM light emission has not yet been directly correlated with the underlying elec-

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